U.S. DEPARTMENT OF COMMERCE
National Technical Information Service
PB-294 793

Thorium Fuel-Cycle Alternatives

California Univ, Berkeley Dept of Nuclear Engineering

Prepared for

Office of Radiation Programs, Las Vegas, NV

Nov 78

United States
Environmental Protection
Agency

Office of Radiation Programs
Las Vegas Facility
P.O. Box 15027
Las Vegas NV 89114

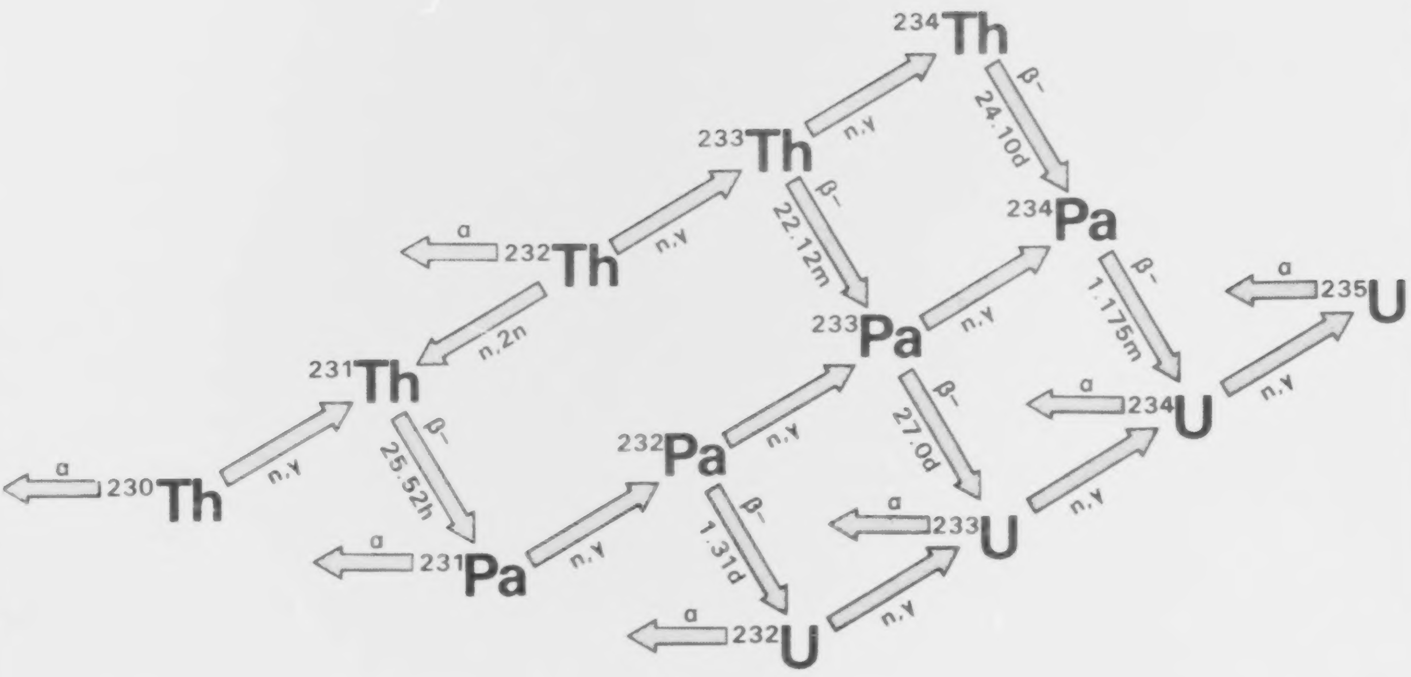
EPA 520/6-78-008
November 1978

PB 294793

Radiation

EPA

Thorium Fuel Cycle Alternatives


^{234}Th
β−
24.10d
n,γ
^{233}Th
β−
22.12m
^{234}Pa
α
^{232}Th
n,γ
β−
1.175m
α
^{235}U
n,2n
^{233}Pa
n,γ
^{231}Th
β−
α
^{234}U
n,γ
β−
25.52h
^{232}Pa
n,γ
27.0d
α
^{230}Th
n,γ
β−
1.31d
α
^{233}U
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^{231}Pa
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α
^{232}U
n,γ

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TECHNICAL REPORT DATA

(Please read Instructions on the reverse before completing)

1. REPORT NO.
EPA 520/6-78-008

2.

3. RECIPIENT'S ACCESSION NO.
PB294793

4. TITLE AND SUBTITLE
Thorium Fuel-Cycle Alternatives

5. REPORT DATE
November 1978

6. PERFORMING ORGANIZATION CODE

7. AUTHOR(S)
T. H. Pigford and C. S. Yang

8. PERFORMING ORGANIZATION REPORT NO.
UCB-NE-3227

9. PERFORMING ORGANIZATION NAME AND ADDRESS
Department of Nuclear Engineering
University of California
Berkeley, California 94720

10. PROGRAM ELEMENT NO.

11. CONTRACT/GRANT NO.
68-01-1962

12. SPONSORING AGENCY NAME AND ADDRESS
Office of Radiation Programs-Las Vegas Facility
U.S. Environmental Protection Agency
P. O. Box 15027
Las Vegas, NV 89114

13. TYPE OF REPORT AND PERIOD COVERED

14. SPONSORING AGENCY CODE
EPA 520/6

15. SUPPLEMENTARY NOTES

16. ABSTRACT

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17. KEY WORDS AND DOCUMENT ANALYSIS

a. DESCRIPTORS	b. IDENTIFIERS/OPEN ENDED TERMS	c. COSATI Field/Group
Nuclear fuel cycles Thorium Radioactive wastes Nuclear electric power generation Radiation hazards Nuclear materials management	light-water/gas-cooled reactors, nuclear fuel processing, nuclear fuel resources, nuclear power economics, nuclear explosive safeguards, nuclear weapons proliferation, nuclear fuel mgmt.strategy	1810 0702 1807 1001 0618, 1312 1809

18. DISTRIBUTION STATEMENT
Release unlimited

19. SECURITY CLASS *(This Report)*
Unclassified

21. NO. OF PAGES
170

20. SECURITY CLASS *(This page)*
Unclassified

22. PRICE
A08-A01

☆ U.S. GOVERNMENT PRINTING OFFICE: 1979-684-317/2130 Region No. 9-1

EPA 520/6-78-008

(UCB-NE-3227)

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Prepared for
U.S. Environmental Protection Agency
under Contract No. 68-01-1962

November, 1978

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FOREWORD

The Office of Radiation Programs carries out a national program designed to evaluate the exposure of man to ionizing and nonionizing radiation, and to promote the development of controls necessary to protect the public health and safety and assure environmental quality.

Part of this program is devoted to an examination of existing and proposed energy technologies with respect to radiological health impacts. In recent years, a number of studies under government and private sponsorship have been made to examine nuclear fuel-cycle alternatives to the uranium-oxide-fueled light-water reactor presently used in the United States. These have been motivated by a number of considerations, among which include a search for ways to extend the nation's fission-fuel resources, as well as the examination of various fuel-cycle alternatives in terms of nuclear explosives safeguards and nuclear weapons proliferation issues.

Thorium-based fuel cycles have the potential for extending nuclear energy resources. However, fuel cycles which utilize thorium may have features which are significantly different from the uranium-oxide light-water-reactor fuel cycle. As new fuel cycles are examined, major consideration must be given to environmental and safety aspects. A first step in this analysis is the development of descriptions of the basic features of proposed and potential fuel cycles, which includes the identification of the various radionuclides associated with these fuel cycles. It then becomes possible to examine environmental control requirements in a preliminary fashion and to make comparisons between alternative fuel cycles on the basis of these requirements.

The present report provides a basic reference document for the above purpose. Comments on this analysis as well as any new information would be welcomed.

W. D. Rowe
Deputy Assistant Administrator
for Radiation Programs

ABSTRACT

Actinide material quantities and lifetime uranium ore requirements are calculated for thorium fuel cycles in pressurized-water reactors, high-temperature gas-cooled reactors, and pressure-tube heavy-water reactors, and are compared with similar quantities for reference uranium-plutonium fueling in light-water reactors and in fast breeders. Flowsheets are presented for national-international fuel cycles for safeguard controls, including dispersed national reactors fueled with thorium and denatured uranium. Long-term radioactivity properties of high-level radioactive wastes are compared. Also compared are the production of ^{14}C, ^{3}H, ^{232}U, and other activated radionuclides from these reactors and fuel cycles.

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EPA 520/6-78-008

UCB-NE 3227

THORIUM FUEL-CYCLE ALTERNATIVES

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1. Introduction

The purpose of this report is to summarize features of alternative power reactor fuel cycles utilizing thorium. This is a follow on to an earlier study, whereby the fuel cycle material quantities and environmental effluents from the thorium-uranium HTGR fuel cycle were analyzed. [a] The objective of the present study [b] is to establish representative material quantities and overall fuel-cycle flowsheets for alternate thorium cycles and to discuss those features of thorium fuel cycles which may be pertinent to future EPA planning concerning the environmental issues of thorium utilization.

Emphasis is given to those fuel cycles which could be implemented in reactors which are already commercialized or which have been brought to the stage of commercial application, i.e., light water reactors (LWR), heavy water reactors (HWR), and the high-temperature gas-cooled reactor (HTGR). Thorium fueling of non-breeder reactors requires a separate fissile make-up, which may be in the form of enriched uranium from isotope separation or plutonium recovered from uranium fueling. Both techniques are considered in the cases studied here. Also, consideration is given to the possible use of thorium as core and/or blanket material in fast-breeder reactors.

Of these reactors, the HTGR is the only one in which thorium has been selected in the reference design. The designer, General Atomic, has concluded that the fuel cycle economics are more favorable for U-Th fueling than for fueling with low-enrichment uranium, even with current and near-term estimates of the costs of uranium ore and of fuel cycle operations. In light-water and heavy-water reactors, current and near-term costs favor uranium fueling, but future higher costs of less concentrated uranium ores may eventually tilt the choice towards thorium.

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[a] The earlier UC study (P1) was carried out under EPA Contract No. 68-01-0561

[b] Carried out under EPA Contract No. 68-01-1962.

The current national interest in thorium fuel cycles is directed towards:

(a) improved utilization of uranium resources

(b) use of thorium with denatured uranium as a possible means of reducing the threat of international proliferation of nuclear explosives.

Since these considerations may strongly influence the choice of a particular fuel cycle, with its concomitant environmental problems, the features of each of these thorium fuel cycles with respect to resource utilization and proliferation are also discussed in this respect.

This report also presents comparisons of the radioactive wastes which result from these fuel cycles, with emphasis upon the actinide content of high-level wastes. Differences in the production rate of tritium, ^{14}C, and other activated species present in discharge fuel-reprocessing wastes are also considered.

The report first establishes, as a basis for comparison, the principal fuel-cycle quantities for uranium fueling in light-water reactors. Possible flowsheets for adapting these light-water reactors to thorium fueling are then described and resource requirements and radioactive waste properties are compared. Similar comparisons to the base case of uranium fueling are made for the heavy-water reactors and HTGR reactors. The results of these comparisons are summarized in Section 10. Details of the computational methods used in the study are summarized in Appendix C.

2. Actinide Reactions

Since most of the important differences in the characteristics of fuel cycles with and without thorium result from differences in actinide composition of the fuel, these differences can best be followed by first examining the actinide reactions in thorium fuel. Actinide reactions for uranium-plutonium fuel have been described elsewhere (P2). The principal actinides involved in using thorium-uranium fuel are shown in the actinide chain of Figure 2.1. The most important reactions are the fission of ^{233}U and ^{235}U and the absorption of neutrons in ^{232}Th to form ^{233}U.

The relatively long 27.0-day half life of ^{233}Pa, the precursor of ^{233}U, may affect the time that irradiated fuel must be stored prior to reprocessing. If the discharged fuel is stored only for 150 days, as is frequently specified for sufficient decay of ^{131}I, some of the ^{233}Pa will remain during reprocessing. Protactinium is one of the most difficult of the elements to separate from uranium, and the high radioactivity of protactinium may contribute to the problem of decontaminating the uranium product after it is separated from the fission products and thorium. Also, for a short period of pre-reprocessing storage, ^{233}Pa would have to be recovered or else its loss would represent an appreciable fissile loss in the fuel cycle. Another effect of the relatively long half life of ^{233}Pa is the build-up of ^{233}U in reactor fuel due to ^{233}Pa decay after shutdown, thereby adding to requirements for reactivity control.

Another problem of the thorium fuel cycle results from the radioactivity of 72-yr ^{232}U and its daughters (B1). ^{232}U is formed by (n,2n) reaction with ^{232}Th according to:

$$^{232}Th \xrightarrow{n,2n} {}^{231}Th \xrightarrow[25.5hr]{\beta-} {}^{231}Pa \xrightarrow{n,\gamma} {}^{232}Pa \xrightarrow[1.32d]{\beta-} {}^{232}U$$

and by

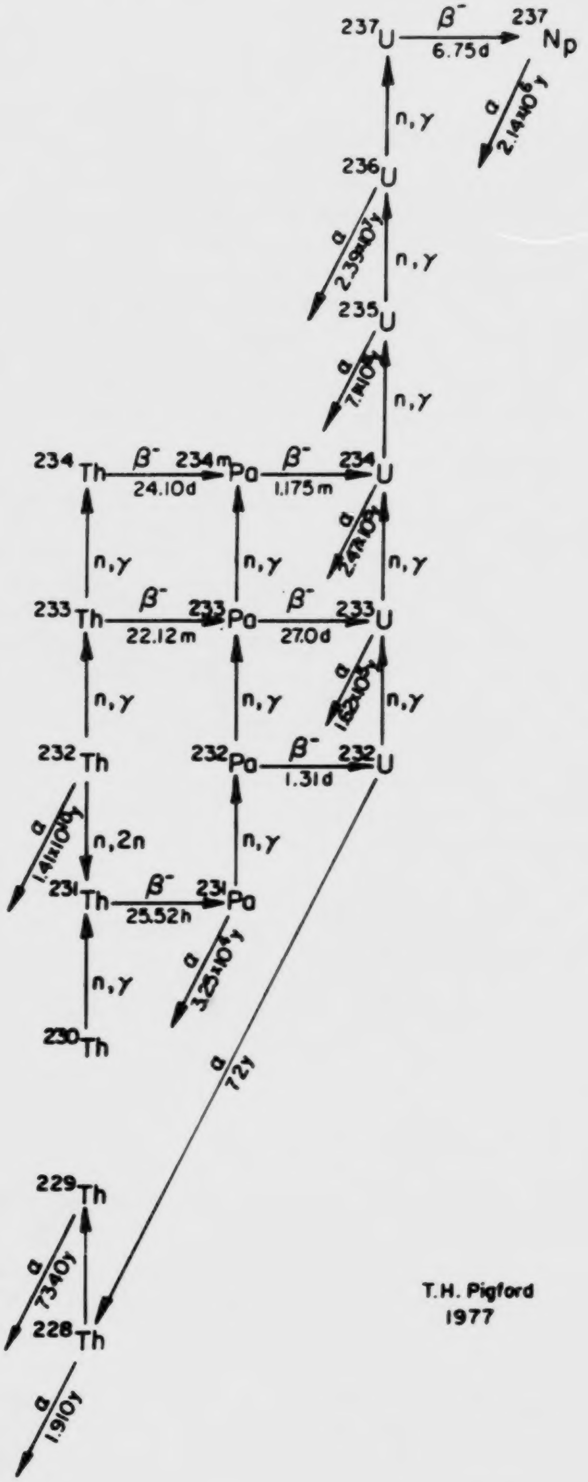
$$^{233}U \xrightarrow{n,2n} {}^{232}U$$

It is also formed by the chain initiating with ^{235}U:

$$^{235}U \xrightarrow{n,\gamma} {}^{236}U \xrightarrow{n,\gamma} {}^{237}U \xrightarrow[6.75d]{\beta-} {}^{237}Np \xrightarrow{n,2n} {}^{236}Np \xrightarrow[22h]{\beta} {}^{236}Pu \xrightarrow[2.85y]{\alpha} {}^{232}U$$

Also, many thorium ores as well as thorium which is obtained as a by-product of uranium mining contain traces of ^{230}Th, a radionuclide in the decay chain of ^{238}U. Neutron absorption in ^{230}Th also results in the formation of ^{232}U:

$$^{230}Th \xrightarrow{n,\gamma} {}^{231}Th \xrightarrow[25.52h]{\beta-} {}^{231}Pa \xrightarrow{n,\gamma} {}^{232}Pa \xrightarrow[1.31d]{\beta-} {}^{232}U$$


237U
β⁻
6.75d
237Np
α
2.14x10⁶y
n,γ
236U
α
2.39x10⁷y
n,γ
235U
α
7.1x10⁸y
n,γ
234Th
β⁻
24.10d
234mPa
β⁻
1.175m
234U
α
2.47x10⁵y
n,γ
n,γ
n,γ
233Th
β⁻
22.12m
233Pa
β⁻
27.0d
233U
α
1.62x10⁵y
n,γ
n,γ
n,γ
232Th
232Pa
β⁻
1.31d
232U
α
1.41x10¹⁰y
n,2n
n,γ
231Th
β⁻
25.52h
231Pa
α
3.25x10⁴y
n,γ
230Th
α
72y
229Th
α
7340y
228Th
α
1.910y

T.H. Pigford
1977

Fig. 2.1 Actinide chains in thorium fuel

Although significant alpha activity results from ^{232}U in the ^{233}U to be recovered and recycled, more of a problem results from the ^{232}U daughters. The ^{232}U decay daughter is 1.91-yr ^{228}Th, a radionuclide which is also formed by the radioactive decay of ^{232}Th. As shown in Figure 2.2, the decay daughters of ^{228}Th are all short-lived, so they reach secular equilibrium with ^{228}Th after a delay time of only a few days. The decay of ^{212}Bi and ^{208}Tl are accompanied by very energetic and penetrating gammas, so gamma shielding is required when fabricating fuel from recycled uranium containing ^{232}U.

Although chemical reprocessing yields essentially pure uranium, storage after separation and time elapsed in shipping to fabrication allow the build-up of ^{228}Th and its decay daughters. Consequently, the gamma activity in separated uranium containing ^{232}U increases continuously with storage time, until it reaches a maximum at about ten years after separation. The calculated growth in activity and gamma dose rate for uranium metal containing 100 ppm ^{232}U is shown in Figure 2.3. As shown later, ^{232}U concentrations in uranium recovered from irradiated thorium may vary from a few hundred to a few thousand parts per million.* Once uranium has been separated from thorium by Thorex partitioning, there is considerable incentive to complete the uranium purification and fuel fabrication quickly to avoid the increasing radiation due to the build up of ^{228}Th. Hydrogenous shielding is also necessary because of the high-energy neutrons from alpha decay in recycled uranium. The alphas from the decay of ^{233}U, ^{232}U, and ^{228}Th interact with light elements such as oxygen and carbon to form neutrons, so the neutron activity also increases with storage time.

The ^{228}Th appearing with the separated thorium results in appreciable radioactivity in the thorium. Consequently, as discussed in Appendix A, it may not be practicable to recycle the recovered thorium until it has been stored for about 3 to 16 yr, depending upon the radioactivity of the uranium with which it is to be used in fuel fabrication.

When ^{235}U is used as fissile make-up in the thorium cycle, as in the reference HTGR fuel cycle, the high burnup and uranium recycle result in considerable production of ^{237}Np, according to the reactions shown in Figure 2.1. Neutron absorption in ^{237}Np then results in a relatively large activity of ^{238}Pu. The plutonium activity is important because of the problems of decontaminating uranium from plutonium when reprocessing the uranium. Also, even though fissile plutonium is formed by neutron absorption in the ^{238}U accompanying the highly

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* Calculated quantities of ^{232}U in various fuel cycles are summarized in Section 9.5 of Chapter 9.

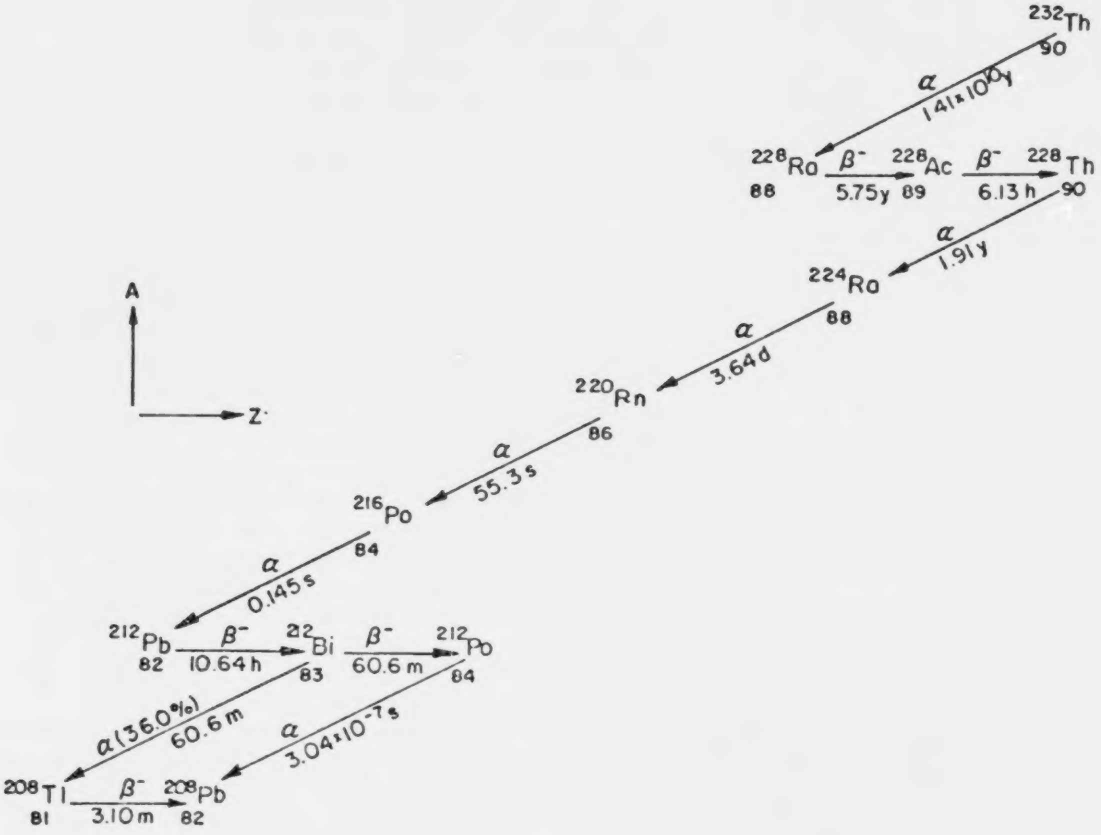
A
Z
^{232}Th 90
α 1.41×10^{10}y
^{228}Ra 88
β$^-$ 5.75y
^{228}Ac 89
β$^-$ 6.13 h
^{228}Th 90
α 1.91y
^{224}Ra 88
α 3.64d
^{220}Rn 86
α 55.3s
^{216}Po 84
α 0.145s
^{212}Pb 82
β$^-$ 10.64h
^{212}Bi 83
β$^-$ 60.6 m
^{212}Po 84
α(36.0%) 60.6m
α 3.04×10^{-7}s
^{208}Tl 81
β$^-$ 3.10m
^{208}Pb 82

Fig. 2.2 Radioactive decay of natural thorium

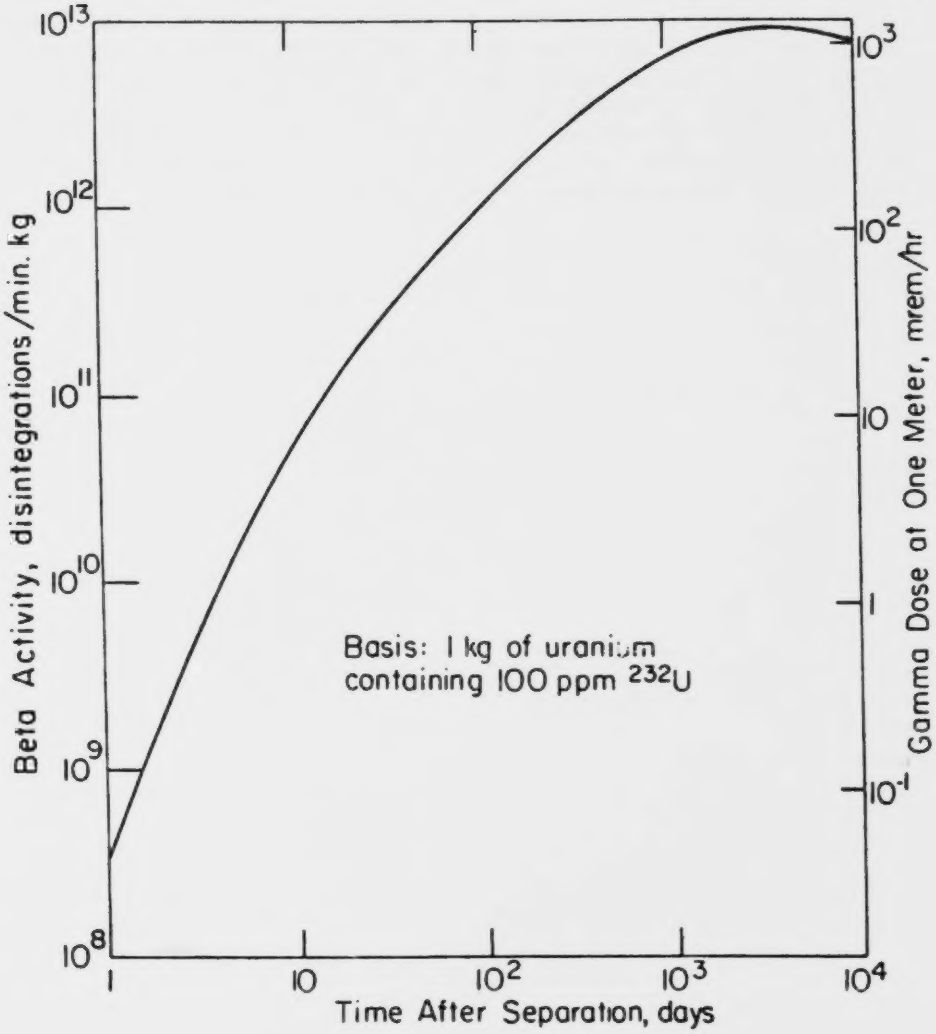

Fig. 2.3 Growth of beta activity and gamma dose due to ^{232}U in Uranium

enriched ^{235}U make-up, as shown in Figure 2.4, the high activity of ^{238}Pu may discourage the utilization of the fuel value of plutonium in the discharge fuel.

When ^{235}U is used as fissile make-up in thorium-uranium fuel, relatively little ^{239}Pu, ^{240}Pu, ^{241}Pu, Am, and Cm are formed. However, when plutonium is used as fissile make-up in a thorium fuel cycle considerable quantities of americium and curium are formed. These are the radionuclides which are the greatest contributors to radioactivity and potential toxicity of the high-level wastes after about 600 years of waste isolation, when most of the fission products have decayed. The effects of actinides upon the long-term radioactivity properties of high-level radioactive wastes from the various fuel cycles are considered in more detail in Chapter 8.

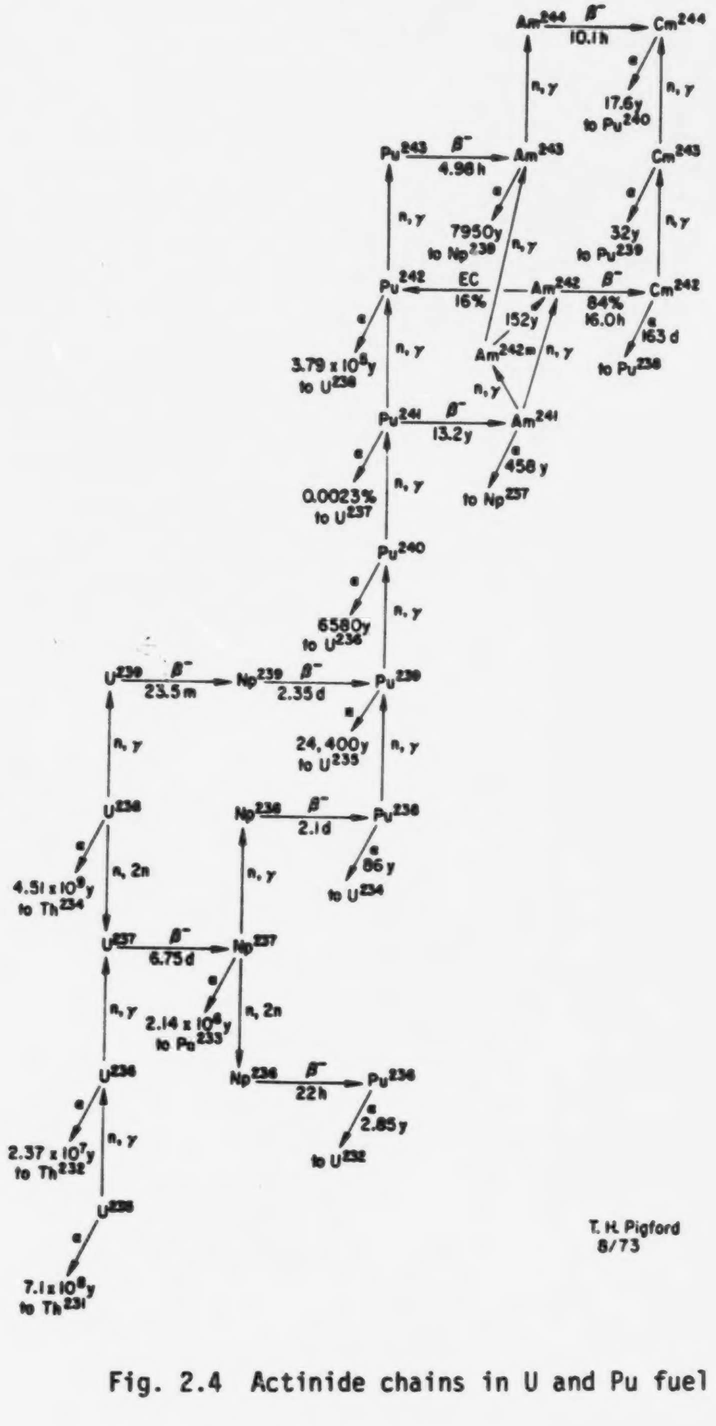
Am^{244}
β^-
10.1h
Cm^{244}
α
17.6y
to Pu^{240}
n, γ
n, γ
Pu^{243}
β^-
4.98 h
Am^{243}
Cm^{243}
n, γ
α
7950y
to Np^{239}
n, γ
α
32y
to Pu^{239}
n, γ
Pu^{242}
EC
16%
Am^{242}
β^-
84%
16.0h
Cm^{242}
α
163 d
to Pu^{238}
α
3.79 x 10^{5}y
to U^{238}
n, γ
152y
Am^{242m}
n, γ
n, γ
Pu^{241}
β^-
13.2y
Am^{241}
α
458 y
to Np^{237}
α
0.0023%
to U^{237}
n, γ
Pu^{240}
α
6580y
to U^{236}
n, γ
U^{239}
β^-
23.5m
Np^{239}
β^-
2.35d
Pu^{239}
α
24, 400y
to U^{235}
n, γ
n, γ
U^{238}
Np^{238}
β^-
2.1d
Pu^{238}
α
86 y
to U^{234}
α
4.51 x 10^{9}y
to Th^{234}
n, 2n
n, γ
U^{237}
β^-
6.75d
Np^{237}
α
2.14 x 10^{6}y
to Pa^{233}
n, 2n
n, γ
U^{236}
Np^{236}
β^-
22h
Pu^{236}
α
2.85y
to U^{232}
α
2.37 x 10^{7}y
to Th^{232}
n, γ
U^{235}
α
7.1 x 10^{8}y
to Th^{231}

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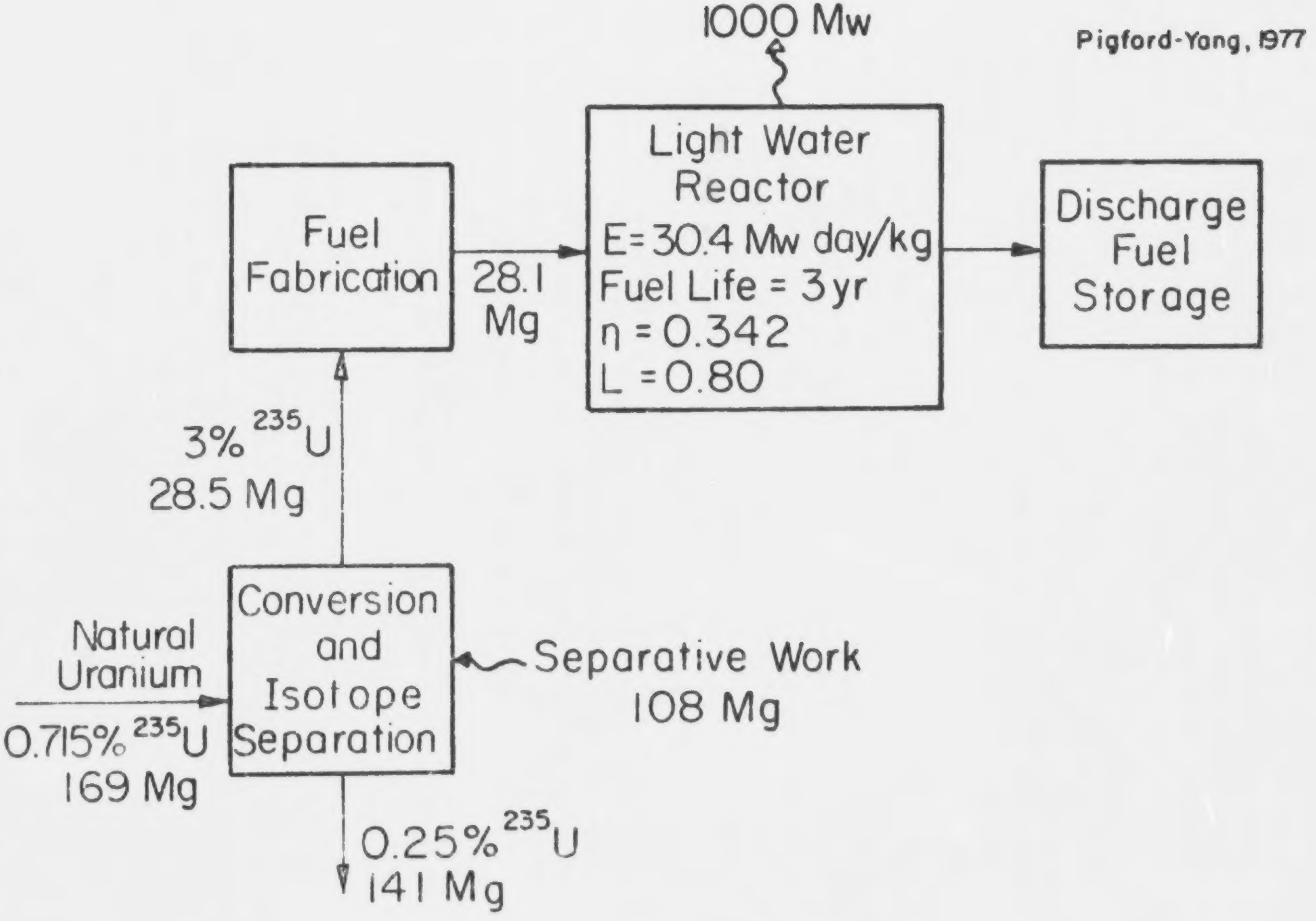
Fig. 2.4 Actinide chains in U and Pu fuel

3. Fuel Cycles for Light-Water Reactors

3.1 Uranium Fueling as a Reference Case

To establish a reference for comparison with thorium cycles, the familiar fuel cycle characteristics of uranium-fueled light-water reactors are illustrated in Figures 3.1, 3.2, 3.3. These are simplified versions of more detailed flowsheets wherein process losses have been taken into account. They have been derived from cycle-by-cycle data calculated by Shapiro et al. (S1) for a pressurized-water reactor. At present in the United States the only operable fuel cycle is the non-reprocessing cycle shown in Figure 3.1, wherein the reactor discharge fuel is stored in water canals. Prior to the administrative decision to defer fuel reprocessing, the fuel cycle involving reprocessing with uranium recycle (Figure 3.2) with storage of the recovered plutonium could have been operable in the U.S. upon completion of the generic licensing decision on reprocessing with fuel recycle, originally scheduled for mid 1977, and upon final licensing of the Barnwell plant, now the only U.S. facility for commercial nuclear fuel processing. Assuming a new and affirmative decision to proceed with reprocessing, recycle of plutonium as well as uranium could not begin until a facility to convert $Pu(NO_3)_4$ to PuO_2 is constructed at the Barnwell reprocessing plant. This would require additional funding and about four years for construction. Reprocessing at Barnwell with uranium recycle and $PU(NO_3)_4$ storage could begin earlier, but it would be limited to about 15 months at full throughput of 1500 Mg/yr, because of limitations on $Pu(NO_3)_4$ storage capacity (C1). Therefore, U.S. reprocessing with both uranium and plutonium recycle does not seem possible until the early 1980's, and it may be deferred beyond that date because of the delays which have been imposed by the federal administration. Thus, it appears that the commercial fuel reprocessing facilities in England and France may continue as the only means of reprocessing power reactor fuel for many years to come. For many years the U.S. reactors will operate on the non-reprocessing cycle, requiring the construction of additional and enlarged facilities for storing discharge fuel at reactor sites as well as centralized discharge-fuel storage facilities.

Even though storage of discharge fuel does not appreciably detract from the economic benefit of nuclear power in the United States (P3), there will remain considerable incentive to proceed with reprocessing. The principal motivations in the U.S. for reprocessing are (1) to achieve the economic benefits from reprocessing and uranium and plutonium recycle, (2) to reduce the required consumption of uranium ore, (3) to reduce the required uranium-enrichment capacity, (4) to provide plutonium when needed to start the breeder reactors, and (5) to obtain additional commercial-scale experience


1000 Mw
Light Water
Reactor
E=30.4 Mw day/kg
Fuel Life = 3yr
η = 0.342
L = 0.80
Fuel
Fabrication
28.1
Mg
Discharge
Fuel
Storage
3% 235U
28.5 Mg
Conversion
and
Isotope
Separation
Natural
Uranium
0.715% 235U
169 Mg
Separative Work
108 Mg
0.25% 235U
141 Mg

Pigford-Yang, 1977

Fig. 3.1 Lifetime-average annual quantities for uranium-fueled PWR with no fuel reprocessing

TABLE 3.1. Actinides in the Fuel Charged to U-Fueled PWR (1000 Mwe, With or Without U Recycle[a])

Radionuclide		kg/yr	Ci/yr	Weight %
Uranium[b]	235	$8.43 \times 10^{+02}$	1.81	3.00
	238	$2.78 \times 10^{+04}$	9.09	97.00
Total		$2.81 \times 10^{+04}$	$\alpha = 1.09 \times 10^{1}$	100.00

[a] 30.4 Mw-day/kg HM, 34.2% thermal efficiency, 80% capacity factor, near-equilibrium fuel cycle.

[b] ^{234}U is not included

TABLE 3.2 . Actinides in the Fuel Discharged from the U-Fueled PWR[a] (1000 Mwe, U Fuel With or Without U Recycle[b])

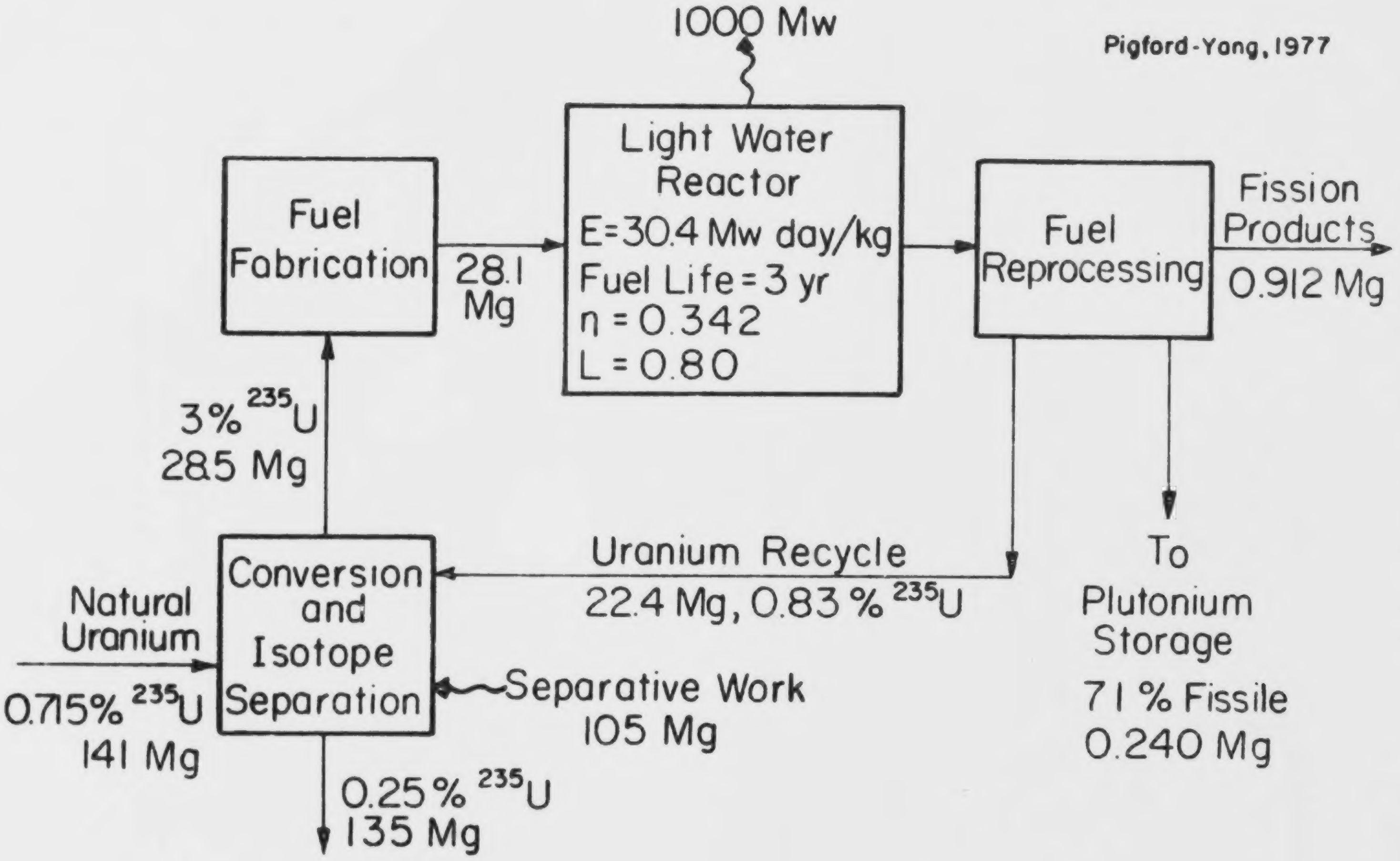
Radionuclide		kg/yr	Ci/yr	Weight %
Uranium[c]	235	$2.25 \times 10^{+02}$	4.82×10^{-01}	0.83
	236	$1.06 \times 10^{+02}$	6.72	0.39
	238	$2.66 \times 10^{+04}$	8.87	98.78
Total		$2.69 \times 10^{+04}$	$\alpha = 1.61 \times 10^{+01}$	100.00
Plutonium[d]	239	$1.40 \times 10^{+02}$	$8.59 \times 10^{+03}$	57.39
	240	$5.70 \times 10^{+01}$	$1.29 \times 10^{+04}$	23.05
	241	$3.50 \times 10^{+01}$	$3.94 \times 10^{+06}$	14.43
	242	$1.20 \times 10^{+01}$	$4.68 \times 10^{+01}$	5.13
Total		$2.44 \times 10^{+02}$	$\alpha = 2.15 \times 10^{+04}$ $\beta = 3.94 \times 10^{+06}$	100.00

[a] immediately after discharge

[b] 30.4 Mw-day/kg HM, 34.2% thermal efficiency, 80% capacity factor, near-equilibrium fuel cycle.

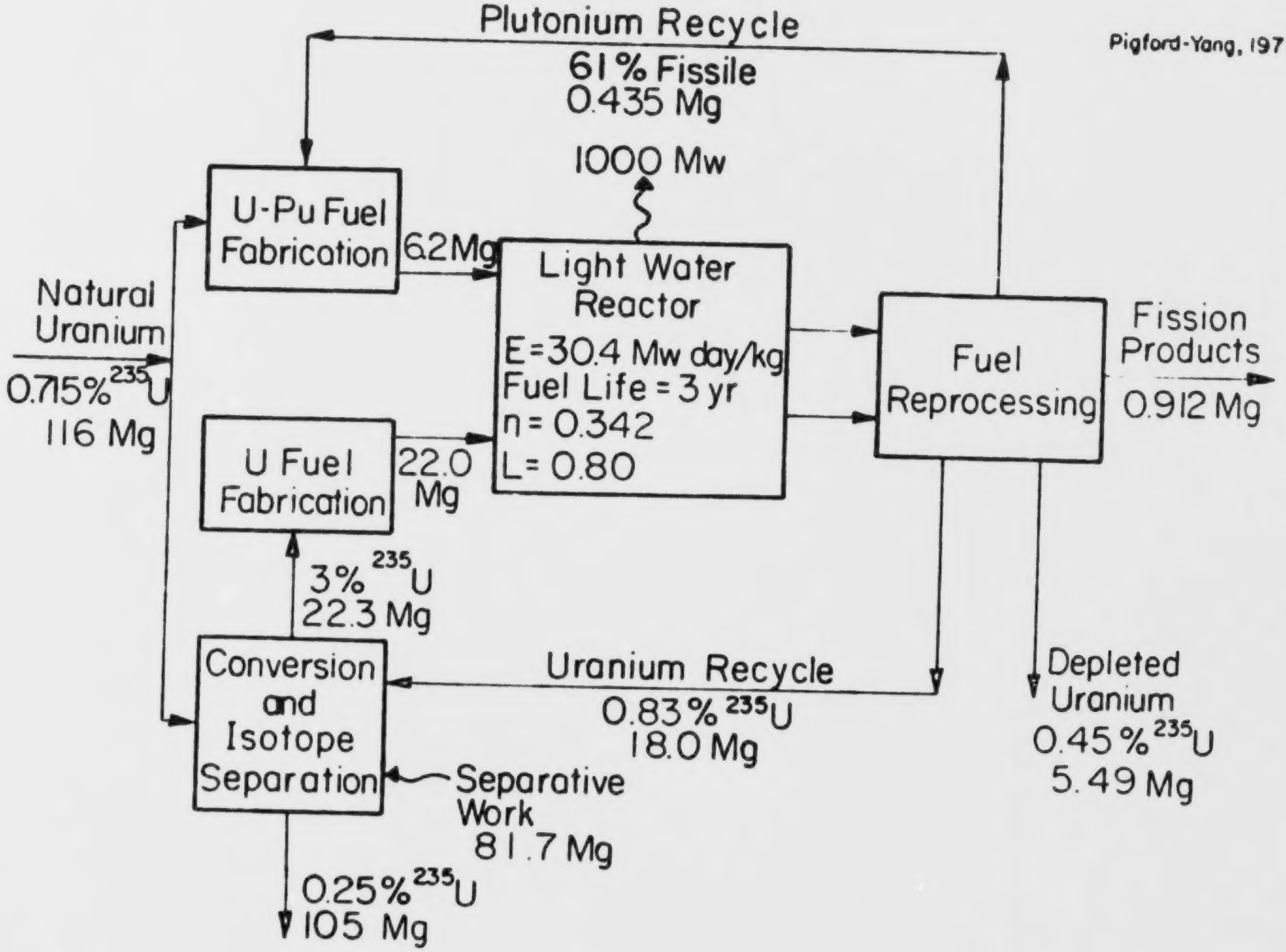
[c] ^{234}U, ^{237}U and ^{239}U are not included

[d] ^{236}Pu, ^{238}Pu, and ^{243}Pu are not included


1000 Mw
Light Water Reactor
E=30.4 Mw day/kg
Fuel Life = 3 yr
η = 0.342
L = 0.80
Fuel Fabrication
28.1 Mg
Fuel Reprocessing
Fission Products
0.912 Mg
3% ^{235}U
28.5 Mg
Conversion and Isotope Separation
Natural Uranium
0.715% ^{235}U
141 Mg
Uranium Recycle
22.4 Mg, 0.83 % ^{235}U
Separative Work
105 Mg
0.25 % ^{235}U
135 Mg
To Plutonium Storage
71 % Fissile
0.240 Mg

Pigford-Yang, 1977

Fig. 3.2 Lifetime-average quantities for uranium-fueled PWR with fuel reprocessing and uranium recycle (E=fuel exposure, η=overall thermal efficiency, L=capacity factor)


Plutonium Recycle
61% Fissile
0.435 Mg
1000 Mw
U-Pu Fuel Fabrication
6.2 Mg
Light Water Reactor
E=30.4 Mw day/kg
Fuel Life = 3 yr
η = 0.342
L = 0.80
Natural Uranium
0.715% 235U
116 Mg
U Fuel Fabrication
22.0 Mg
Fuel Reprocessing
Fission Products
0.912 Mg
3% 235U
22.3 Mg
Conversion and Isotope Separation
Uranium Recycle
0.83% 235U
18.0 Mg
Depleted Uranium
0.45% 235U
5.49 Mg
Separative Work
81.7 Mg
0.25% 235U
105 Mg

Pigford-Yang, 1977

Fig. 3.3 Lifetime-average annual quantities for uranium-fueled PWR with self-generated plutonium recycle (E=fuel exposure, η= overall thermal efficiency, L=capacity factor)

TABLE 3.3. Actinides in the Fuel Charged to the PWR with Self-Generated Pu Recycle (1000 Mwe, with U and Pu Recycle[a])

1. 3% ^{235}U make-up fuel

Radionuclide		kg/yr	Ci/yr	Weight %
Uranium[b]	235	$5.97 \times 10^{+02}$	1.28	3.00
	238	$1.93 \times 10^{+04}$	6.43	97.00
Total		$1.99 \times 10^{+04}$	7.71	100.00

2. Recycled plutonium fuel[c]

Radionuclide		kg/yr	Ci/yr	Weight %
Uranium[b]	235	$5.40 \times 10^{+01}$	1.16×10^{-01}	0.71
	238	$7.49 \times 10^{+03}$	2.50	99.29
Total		$7.54 \times 10^{+03}$	2.62	100.00
Plutonium[d]	239	$2.66 \times 10^{+02}$	$1.63 \times 10^{+04}$	40.00
	240	$1.98 \times 10^{+02}$	$4.49 \times 10^{+04}$	29.78
	241	$1.17 \times 10^{+02}$	$1.32 \times 10^{+07}$	17.59
	242	$8.40 \times 10^{+01}$	$3.28 \times 10^{+02}$	12.63
		$6.65 \times 10^{+02}$	$\alpha = 6.15 \times 10^{+04}$	100.00
			$\beta = 1.32 \times 10^{+07}$	

[a] 30.4 Mw-day/kg HM, 34.2% thermal efficiency, 80% capacity factor, near-equilibrium cycle.

[b] ^{234}U is not included.

[c] 150 days cooling of discharged fuel before reprocessing. 1.5% loss in reprocessing, 1.5% loss in fabrication. Natural uranium is added to the recycled plutonium to dilute the recycle fuel to proper enrichment.

[d] ^{236}Pu and ^{238}Pu are not included.

TABLE 3.4. Actinides in the Fuel Discharged From The PWR with Self-Generated Pu Recycle[a] (1000 Mwe, with U and Pu Recycle[b])

1. 3% ^{235}U make-up fuel

Radionuclide		kg/yr	Ci/yr	Weight %
Uranium[c]	235	$1.59 \times 10^{+2}$	3.41×10^{-1}	0.83
	236	$7.50 \times 10^{+1}$	4.76	0.39
	238	$1.89 \times 10^{+4}$	6.28	98.78
Total		$1.91 \times 10^{+04}$	$\alpha = 1.14 \times 10^{+01}$	100.00
Plutonium[d]	239	$9.90 \times 10^{+01}$	$6.07 \times 10^{+03}$	57.39
	240	$4.10 \times 10^{+01}$	$9.29 \times 10^{+03}$	23.05
	241	$2.50 \times 10^{+01}$	$2.81 \times 10^{+06}$	14.43
	242	9.00	$3.51 \times 10^{+01}$	5.13
Total		$1.74 \times 10^{+02}$	$\alpha = 1.54 \times 10^{+04}$ $\beta = 2.81 \times 10^{+06}$	100.00

2. Recycled plutonium fuel

Radionuclide				
Uranium[c]	235	$3.40 \times 10^{+01}$	7.29×10^{-02}	0.47
	236	4.00	2.54×10^{-01}	0.06
	238	$7.34 \times 10^{+03}$	2.45	99.47
Total		$7.38 \times 10^{+03}$	$\alpha = 2.78$	100.00
Plutonium[d]	239	$1.85 \times 10^{+02}$	$1.13 \times 10^{+04}$	34.42
	240	$1.70 \times 10^{+02}$	$3.85 \times 10^{+04}$	31.83
	241	$9.80 \times 10^{+01}$	$1.10 \times 10^{+07}$	18.31
	242	$8.20 \times 10^{+01}$	$3.20 \times 10^{+02}$	15.44
Total		$5.35 \times 10^{+02}$	$\alpha = 5.01 \times 10^{+04}$ $\beta = 1.10 \times 10^{+07}$	100.00

[a] immediately after discharge

[b] 30.4 Mw-day/kg HM, 34.2% efficiency, 80% capacity factor, near equilibrium fuel cycle

[c] ^{234}U, ^{237}U and ^{239}U are not included

[d] ^{236}Pu, ^{238}Pu, and ^{243}Pu are not included

on fuel reprocessing so as to provide better foundation for future facilities necessary to reprocess discharge fuel from breeders. Similar reprocessing will also be necessary to obtain any significant benefits from thorium fueling.

The uranium-plutonium recycle flowsheet of Figure 3.3 is calculated for self-generated plutonium recycle. Alternatively, the plutonium recovered from fuel discharged from uranium-fueled reactors can be blended with natural uranium to refuel another light-water reactor, as shown in Figure 3.4. This uranium-plutonium fueled reactor is similar to the uranium-fueled reactor in Figure 3.1, but it requires a larger number of control absorbers because of the large neutron-reaction cross section of fissile plutonium. Additional plutonium is formed in this reactor during irradiation, and the plutonium in the discharge fuel is recovered and recycled. The make-up plutonium for this cycle very nearly equals that recovered from the fuel discharged by about three uranium-fueled reactors operating as in Figure 3.2.

Such uranium-plutonium-fueled water reactors have been suggested as plutonium-burner reactors to be located at centers where are also co-located facilities for fuel reprocessing and for fabricating recycle uranium-plutonium fuel. Discharge fuel from externally located uranium-fueled reactors fueled with uranium or with uranium and thorium would be sent to these centers for reprocessing, and the recovered plutonium would be consumed on site in the plutonium-burner reactors. Such centers have been proposed as a means of localizing the use of plutonium, thereby avoiding the safeguards issues associated with shipping plutonium (U1, W1). Examples of such international centers servicing off-site national reactors are given in Chapter 7.

Although the uniformity of fuel charged to plutonium-burner reactors should lead to more optimum core loadings for these reactors and greatest economy of plutonium utilization, the first plutonium recovered for recycling is more likely to be returned as a partial reload for the same reactor from which it was discharged. This is the self-generated recycle operation of Figure 3.3. Approximately one fourth of the reactor is fueled with natural uranium blended with recycled plutonium, and three fourths is fueled with uranium enriched to 3% ^{235}U.

3.2 Thorium-Fueled Pressurized-Water Reactors

The same pressurized-water reactor can be adapted to thorium fueling, whereby natural ^{232}Th replaces the function of the ^{238}U isotope in the previous flowsheets. The make-up fissile material is either 93.5% ^{235}U, as shown in Figure 3.5, or plutonium recovered from the discharge fuel from uranium-fueled water reactors, as shown

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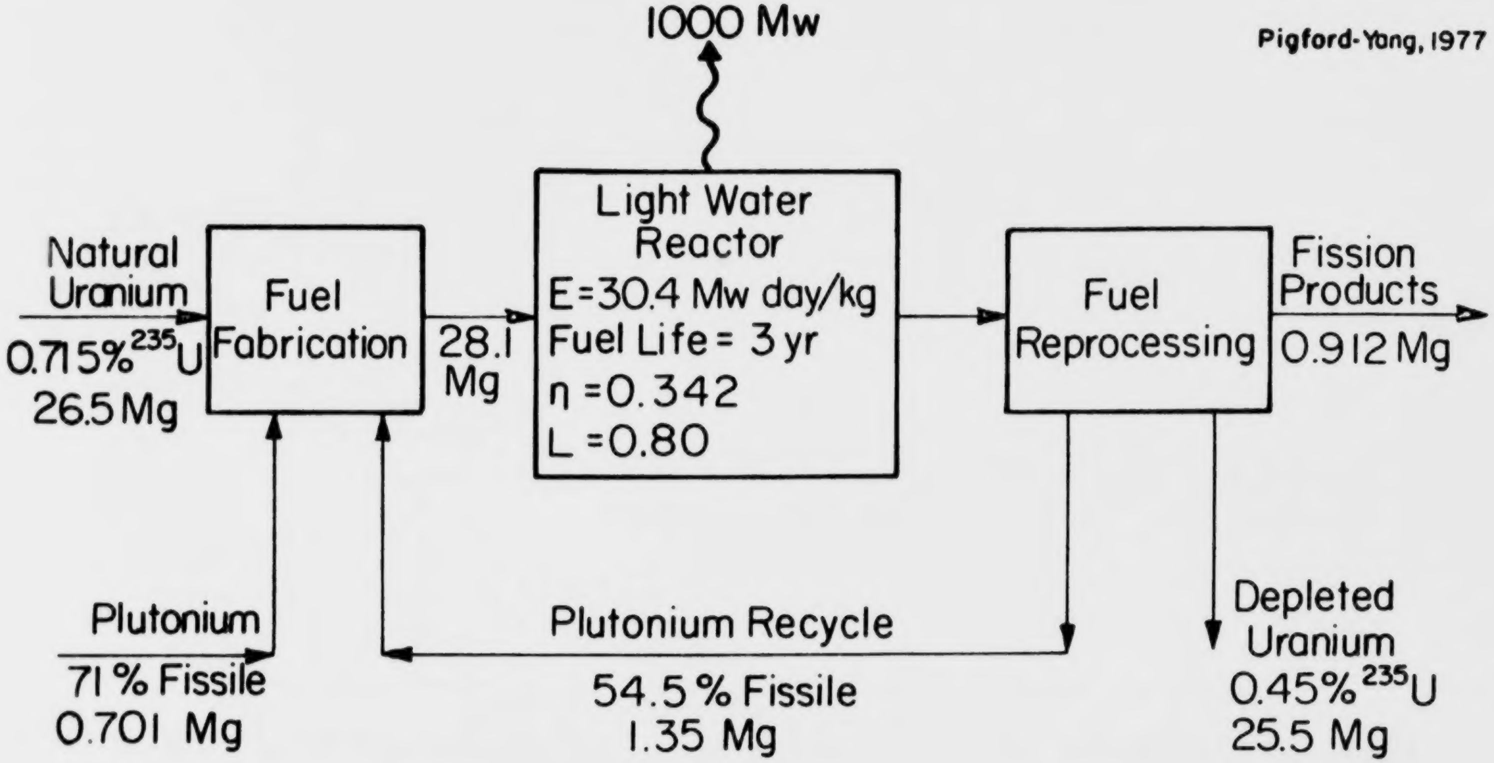
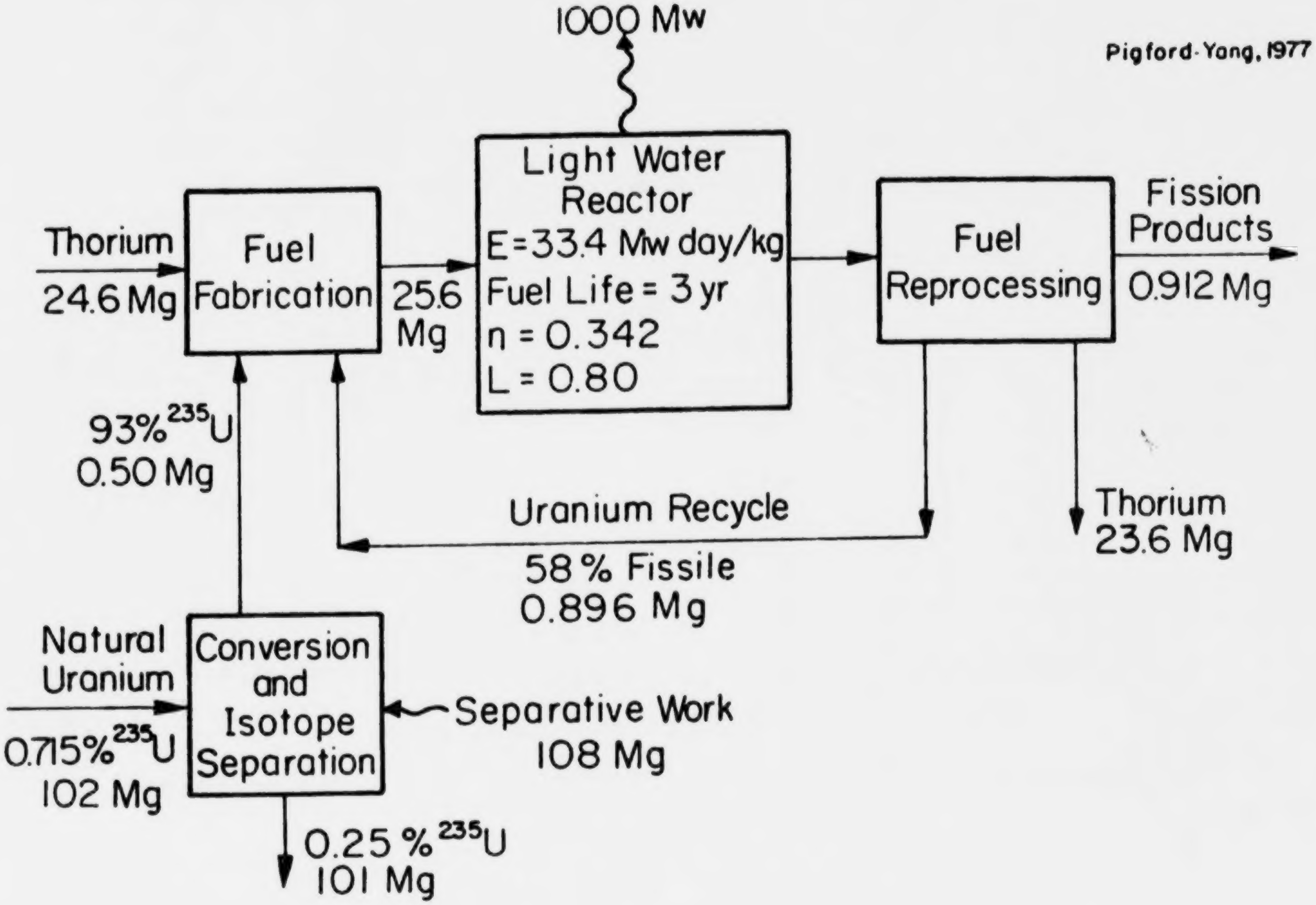

1000 Mw
Natural Uranium
0.715% ^{235}U
26.5 Mg
Fuel Fabrication
28.1 Mg
Light Water Reactor
E=30.4 Mw day/kg
Fuel Life = 3 yr
η =0.342
L =0.80
Fuel Reprocessing
Fission Products
0.912 Mg
Plutonium
71 % Fissile
0.701 Mg
Plutonium Recycle
54.5 % Fissile
1.35 Mg
Depleted Uranium
0.45% ^{235}U
25.5 Mg

Fig. 3.4 Lifetime-average annual quantities for PWR fueled with plutonium and natural uranium (E=fuel exposure, η=overall thermal efficiency, L=capacity factor)


1000 Mw
Thorium
24.6 Mg
Fuel Fabrication
25.6 Mg
Light Water Reactor
E=33.4 Mw day/kg
Fuel Life = 3 yr
n = 0.342
L = 0.80
Fuel Reprocessing
Fission Products
0.912 Mg
93% ^{235}U
0.50 Mg
Uranium Recycle
58 % Fissile
0.896 Mg
Thorium
23.6 Mg
Natural Uranium
0.715% ^{235}U
102 Mg
Conversion and Isotope Separation
Separative Work
108 Mg
0.25 % ^{235}U
101 Mg

Pigford-Yang, 1977

Fig. 3.5 Lifetime-average annual quantities for PWR fueled with uranium and thorium (E=fuel exposure, η=overall thermal efficiency, L=capacity factor)

in Figure 3.6. The ^{233}U, resulting from neutron absorption in thorium, and other uranium isotopes are recycled. To simplify comparison with the uranium-fueled PWR's discussed in Section 3.1, the same total heat generation per fuel rod for fueling with urania or thoria has been assumed (S1). Because the thoria is of lower density than urania, the average thermal exposure of 30.4 Mw day/kg for urania fuel is equivalent on this basis to 33.4 Mw day/kg for thoria fuel. The recovered thorium is radioactive because of 1.91-yr ^{228}Th and must be stored for several years before it can be recycled (see Appendix A). Detailed data on composition of charge and discharge fuel for the near-equilibrium fuel cycle are given in Tables 3.5 through 3.8. These data indicate a concentration of ^{232}U in the recycled uranium as high as 2600 ppm for the near-equilibrium fuel cycle. This is 3.5 times greater than the current estimate of 742 ppm for ^{232}U in recycled bred uranium for a near-equilibrium fuel cycle for the HTGR, as shown in Chapter 5. Evidently the greater fuel lumping and close-packed lattice of the PWR result in a higher flux of neutrons at energies above the ^{232}Th (n,2n) threshold.

3.3 Resource Requirements for Pressurized-Water Reactors

One purpose for considering thorium cycles in light water reactors is to reduce the demands for uranium ore. The total ore required to fuel a given reactor over its operating life must include the ore to supply the start-up fuel inventory as well as the annual replacement requirements accumulated over the operating life. The lifetime ore requirements calculated for the light-water reactor fuel cycles appear in Table 3.9. For those cases involving recycle of fissile material, and/or supply of fissile material recovered from fuel reprocessing, the reactor is assumed to be fueled with slightly enriched (3%) uranium or with ^{235}U-Th fuel until sufficient fissile inventory is accumulated within the reactor and fuel cycle so that the reactor can then operate on the equilibrium fuel cycle. Therefore, for first-generation recycle reactors there can be a considerable delay before the resource advantage of recycle is manifested. For subsequent reactors operating on the same fuel cycle, i.e. "second-generation" reactors, the accumulated fissile inventory in the core and in the fuel cycle from decommissioned reactors can be taken over so that these new reactors can operate on the equilibrium fuel cycle over their entire operating life, thereby achieving greater ore savings. This assumes, of course, that at the time the "second-generation" light-water reactors are to be constructed there are no new types of reactors, such as breeders, which are more resource efficient and which could better utilize the accumulated fissile inventory.

It is apparent that thorium fueling in water reactors results in only a small reduction in the uranium ore requirements, as

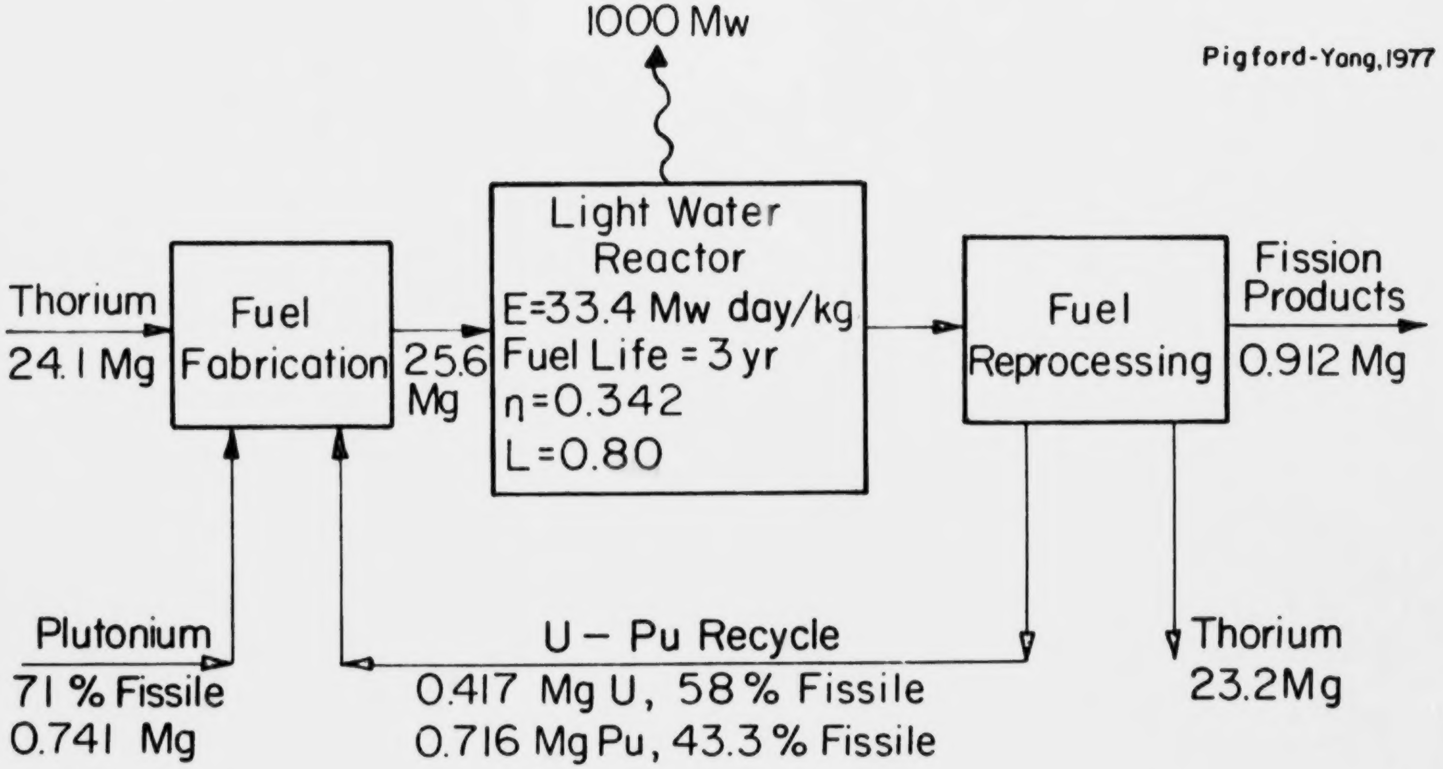

1000 Mw
Pigford-Yang, 1977
Thorium
24.1 Mg
Fuel
Fabrication
25.6
Mg
Light Water
Reactor
E=33.4 Mw day/kg
Fuel Life = 3 yr
η=0.342
L=0.80
Fuel
Reprocessing
Fission
Products
0.912 Mg
Plutonium
71 % Fissile
0.741 Mg
U – Pu Recycle
0.417 Mg U, 58 % Fissile
0.716 Mg Pu, 43.3 % Fissile
Thorium
23.2Mg

Fig. 3.6 Lifetime-average annual quantities for PWR fueled with thorium, plutonium, and recycled uranium (E=fuel exposure, η=overall thermal efficiency, L=capacity factor)

TABLE 3.5. Actinides in the Fuel Charged to the U-Th Fueled PWR (1000 Mwe, with U Recycle[a])

1. 93% ^{235}U and thorium make-up fuel

Radionuclide		kg/yr	Ci/yr	Weight %
Thorium	232	$8.73 \times 10^{+03}$	9.55×10^{-01}	100.00
Total		$8.73 \times 10^{+03}$	$\alpha = 9.55 \times 10^{-01}$	100.00
Uranium	234	$1.90 \times 10^{+01}$	$1.18 \times 10^{+02}$	4.70
	235	$3.67 \times 10^{+02}$	7.87×10^{-01}	93.00
	236	9.00	3.00×10^{-03}	2.30
		$3.95 \times 10^{+02}$	$\alpha = 1.19 \times 10^{+02}$	100.00

2. Recycled uranium fuel[b]

Radionuclide		kg/yr	Ci/yr	Weight %
Thorium	232	$1.53 \times 10^{+04}$	1.67	100.00
Total		$1.53 \times 10^{+04}$	$\alpha = 1.67$	100.00
Uranium	232	1.88	$4.03 \times 10^{+04}$	0.16
	233	$4.34 \times 10^{+02}$	$4.11 \times 10^{+03}$	36.97
	234	$2.16 \times 10^{+02}$	$1.34 \times 10^{+03}$	18.47
	235	$1.75 \times 10^{+02}$	3.75×10^{-01}	14.91
	236	$3.14 \times 10^{+02}$	$1.99 \times 10^{+01}$	26.75
	238	$3.30 \times 10^{+01}$	1.10×10^{-02}	2.81
Total		$1.17 \times 10^{+03}$	$\alpha = 4.58 \times 10^{+04}$	100.00

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[a] 33.4 Mw-day/kg, 34.2% thermal efficiency, 80% capacity factor, near-equilibrium fuel cycle.

[b] 150 days cooling of discharged fuel before reprocessing, 1.5% loss in fabrication. Natural uranium is added to the recycled plutonium to dilute the recycle fuel to the proper enrichment.

TABLE 3.6. Actinides in the Fuel Discharged From the U-Th Fueled PWR[a] (1000 Mwe, with U Recycle[b])

1. U and thorium make-up fuel

Radionuclide		kg/yr	Ci/yr	Weight %
Thorium[c]	232	$8.50 \times 10^{+03}$	9.30×10^{-01}	100.00
	Total	$8.50 \times 10^{+03}$	$\alpha = 9.30 \times 10^{-01}$	100.00
Protactinium[d]	233	$1.20 \times 10^{+01}$	$2.49 \times 10^{+08}$	100.00
	Total	$1.20 \times 10^{+01}$	$\beta = 2.49 \times 10^{+08}$	100.00
Uranium[e]	232	3.40×10^{-01}	$7.28 \times 10^{+03}$	0.12
	233	$1.08 \times 10^{+02}$	$1.02 \times 10^{+03}$	36.94
	234	$2.60 \times 10^{+01}$	$1.61 \times 10^{+02}$	8.89
	235	$1.04 \times 10^{+02}$	$2.23 \times 10^{+01}$	35.58
	236	$4.60 \times 10^{+01}$	2.92	15.73
	238	8.00	2.67×10^{-03}	2.74
	Total	$2.92 \times 10^{+02}$	$\alpha = 8.46 \times 10^{+03}$	100.00

2. Recycled uranium fuel

Radionuclide		kg/yr	Ci/yr	Weight %
Thorium[c]	232	$1.49 \times 10^{+04}$	1.63	100.00
	Total	$1.49 \times 10^{+04}$	$\alpha = 1.63$	100.00
Protactinium[d]	233	$2.00 \times 10^{+01}$	$4.15 \times 10^{+08}$	100.00
		$2.00 \times 10^{+01}$	$\beta = 4.15 \times 10^{+08}$	100.00
Uranium[e]	232	1.61	$3.45 \times 10^{+04}$	0.17
	233	$3.00 \times 10^{+02}$	$2.84 \times 10^{+03}$	32.17
	234	$2.10 \times 10^{+02}$	$1.30 \times 10^{+03}$	22.52
	235	$8.90 \times 10^{+01}$	1.91×10^{-01}	9.54
	236	$3.05 \times 10^{+02}$	$1.93 \times 10^{+01}$	32.70
	238	$2.70 \times 10^{+01}$	9.00×10^{-03}	2.90
	Total	$9.33 \times 10^{+02}$	$\alpha = 3.87 \times 10^{+04}$	100.00

---

[a] immediately after discharge

[b] 33.4 Mw-day/kg HM, 34.2% thermal efficiency, 80% capacity factor, near-equilibrium fuel cycle. Np, Pu, Am, Cm are not included.

[c] Trace quantities of other thorium isotopes are not included.

[d] Trace quantities of other protactinium isotopes are not included.

[e] ^{237}U and ^{239}U are not included.

TABLE 3.7. Actinides in the Fuel Charged to the Pu-U-Th Fueled PWR (1000 Mwe, with U and Pu Recycle[a])

1. Plutonium and thorium fuel

Radionuclide	kg/yr	Ci/yr	Weight %
Thorium 232	$1.13 \times 10^{+04}$	1.24	100.00
Total	$1.13 \times 10^{+04}$	$\alpha = 1.24$	100.00
Make-up Plutonium[b] 239	$3.02 \times 10^{+02}$	$1.85 \times 10^{+04}$	57.42
240	$1.21 \times 10^{+02}$	$2.74 \times 10^{+04}$	23.00
241	$7.60 \times 10^{+01}$	$8.55 \times 10^{+06}$	14.45
242	$2.70 \times 10^{+01}$	$1.05 \times 10^{+02}$	5.13
Total	$5.26 \times 10^{+02}$	$\alpha = 4.60 \times 10^{+04}$ $\beta = 8.55 \times 10^{+06}$	100.00
Recycled Plutonium[b], [c] 239	$2.00 \times 10^{+02}$	$1.23 \times 10^{+04}$	18.82
240	$4.12 \times 10^{+02}$	$9.33 \times 10^{+04}$	36.76
241	$2.22 \times 10^{+02}$	$2.50 \times 10^{+07}$	20.88
242	$2.29 \times 10^{+02}$	$8.93 \times 10^{+02}$	21.54
	$1.06 \times 10^{+03}$	$\alpha = 1.06 \times 10^{+05}$ $\beta = 2.50 \times 10^{+07}$	100.00

2. Recycled uranium fuel[c], [d]

Radionuclide	kg/yr	Ci/yr	Weight %
Thorium 232	$1.21 \times 10^{+04}$	1.32	100.00
Total	$1.21 \times 10^{+04}$	$\alpha = 1.32$	100.00
Uranium 232	1.49	$3.19 \times 10^{+04}$	0.26
233	$3.75 \times 10^{+02}$	$3.55 \times 10^{+03}$	65.62
234	$1.44 \times 10^{+02}$	$8.91 \times 10^{+02}$	25.20
235	$3.40 \times 10^{+01}$	$7.29 \times 10^{+02}$	5.95
236	$1.70 \times 10^{+01}$	1.08	2.97
Total	$5.71 \times 10^{+02}$	$\alpha = 3.63 \times 10^{+04}$	100.00

---

[a] 33.4 Mw-day/kg HM, 34.2% thermal efficiency, 80% capacity factor, near-equilibrium fuel cycle.

[b] ^{236}Pu and ^{238}Pu are not included.

[c] 150 days cooling of recycled fuel before reprocessing. 1.5% loss in reprocessing, 1.5% loss in fabrication.

[d] thorium is added to the recycled uranium to dilute the recycle fuel to proper enrichment.

TABLE 3.8. Actinides in the Fuel Discharged from the Pu-U-Th Fueled PWR[a]

(1000 Mwe, with U and Pu recycle[b])

1. Plutonium-thorium fuel

Radionuclide		kg/yr		Ci/yr	Weight%
Thorium[c]	232	$1.11x10^{+04}$		1.22	100.00
Total		$1.11x10^{+04}$	α =	1.22	100.00
Protactinium[d]		9.00		$5.12x10^{+05}$	100.00
Total		9.00	β =	$5.12x10^{+05}$	100.00
Uranium[e]	232	$2.90x10^{-01}$		$6.21x10^{+03}$	0.20
	233	$1.34x10^{+02}$		$1.27x10^{+03}$	92.23
	234	9.00		$5.57x10^{+01}$	6.19
	235	2.00		$4.29x10^{-03}$	1.38
Total		$1.45x10^{+02}$	α =	$7.36x10^{+03}$	100.00
Plutonium[f]	239	$2.06x10^{+02}$		$1.26x10^{+04}$	18.80
	240	$4.24x10^{+02}$		$9.60x10^{+04}$	38.69
	241	$2.30x10^{+02}$		$2.59x10^{+07}$	20.99
	242	$2.36x10^{+02}$		$9.20x10^{+02}$	21.52
Total		$1.10x10^{+03}$	α =	$1.10x10^{+05}$	100.00
			β =	$2.59x10^{+07}$	

2. Recycled uranium fuel

Radionuclide		kg/yr		Ci/yr	Weight
Thorium[c]	232	$1.17x10^{+04}$		1.28	100.00
Total		$1.17x10^{+04}$	α =	1.28	100.00
Protactinium[d]	233	$1.70x10^{+01}$		$3.53x10^{+08}$	100.00
Total		$1.70x10^{+01}$	β =	$3.53x10^{+08}$	100.00
Uranium[e]	232	1.25		$2.68x10^{+04}$	0.28
	233	$2.35x10^{+02}$		$2.23x10^{+03}$	53.14
	234	$1.47x10^{+02}$		$9.10x10^{+02}$	33.24
	235	$3.60x10^{+01}$		$7.72x10^{-02}$	8.14
	236	$2.30x10^{+01}$		1.46	5.20
Total		$4.42x10^{+02}$	α =	$2.99x10^{+04}$	100.00

a/ immediately after discharge
b/ 33.4 Mw-day/kg HM, 34.2% thermal efficiency, 80% capacity factor, near equilibrium fuel cycle
c/ Trace quantities of other thorium isotopes are not included.
d/ Trace quantities of other protactinium isotopes are not included.
e/ ^{236}U, ^{237}U and ^{238}U are not included
f/ ^{236}Pu and ^{238}Pu are not included

TABLE 3.9 30-Year Lifetime Ore Requirements for Pressurized-Water Reactors (1000 Mwe Electrical Power, 80% Capacity Factor)

	Fuel Cycle	Conversion Ratio	Natural Uranium[a], Metric tons (Mg)		Natural Uranium relative to no recycle[b]	Thorium Metric tons (Mg)[c]
			0.2% depleted U	0.25% depleted U		
(a)	No recycle	0.60	4940	5370	1.00	
(b)	U recycle	0.60	4070	4487	0.84	
(c)	U-Pu self-generated recycle	0.61	3340	3680	0.68	
(d)	^{235}U-Th, U-recycle	0.66	2810	3060	0.57	774
(e)	Pu-Th, U-Pu recycled[d]	0.61	3250	3584	0.67	157
(f)	Second generation Pu-Th, U-Pu recycle[e].	0.61	2560	2790	0.52	220

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[a] Uranium contained in U_3O_8 concentrate. To obtain short tons of U_3O_8 multiply by 1.297.

[b] Calculated for 0.25% depleted U from isotope separation.

[c] Thorium contained in ThO_2 concentrate. To obtain short tons of ThO_2 multiply by 1.2517.

[d] Includes U-fueled water reactor to supply make-up Pu. Total system operates for 30 yr at 1000 Mw.

[e] Starts with equilibrium reactor and fuel-cycle fissile inventory.

compared with the analogous case of uranium fueling with recycle of uranium and plutonium. The greatest ore saving per unit of total generated electrical energy for first-generation thorium fueling results from the ^{235}U-Th system of Figure 3.5. However, the use of fully enriched (93%) ^{235}U is subject to special safeguards concerns, as is discussed in Chapter 6. If a stockpile of discharge fuel from uranium-fueled water reactors were accumulated and then later reprocessed, use of the recovered plutonium to fuel the thorium flowsheet of Figure 3.10 could be a more direct way of initiating thorium fueling. Variations of these thorium cycles which have been suggested to mitigate the safeguards issues are discussed in Chapter 6.

Recent estimates (S1) indicate about the same fuel-cycle costs with Pu-Th fueling as for U fueling with U-Pu recycle. ^{235}U fueling is estimated to be more expensive, because of the relatively high costs of 93% ^{235}U. However, there is too much uncertainty in the cost of fuel reprocessing, particularly for thorium fuel reprocessing, for the small differences in the estimated costs for these fuel cycles to be significant. Also, Thorex reprocessing technology has not been brought to the commercial scale of Purex reprocessing, and additional costs of development can be expected (H1, D5).

Assuming no significant cost advantage for thorium fueling in the near future, thorium fueling could become more attractive at a future time when uranium supplies are more limited and the cost of uranium is relatively high. Then reactors with less uranium consumption would have a greater economic advantage and would be more useful to the power economy. There would also be greater incentive to redesign light water reactors to higher conversion ratios for better ore utilization than is indicated in Table 3.9.

The proposed light-water breeder reactor is such a concept. It involves a higher ratio of fuel to water than in present reactors, separated and localized regions of fissile and fertile material, and the use of moveable fuel for reactivity control. All of these modifications increase neutron absorption in thorium, resulting in higher conversion ratio. Thorium-cycle conversion ratios of near unity seem achievable. However, the breeding gain is very small and specific power is low, so pre-breeders of intermediate conversion ratios are proposed as a means of providing the start-up fissile inventory. Analyses have indicated an increased ore requirement during the period of introducing prebreeders and breeders, and net ore savings only after a very long period of operation. Thus, the value of the light-water breeder is marginal relative to other alternatives.

A possibly more useful and realistic concept, to improve the conversion ratio and ore consumption with near-term light-water reactors, is to modify these reactors for "spectral shift" operation (B2). The reactor coolant system could be modified so that heavy water (D_2O) could be introduced into the coolant at controlled concentrations. After each refueling cycle the excess neutron production from fresh fuel would be controlled by replacing enough H_2O with D_2O for less efficient neutron moderation. This excess neutron production, normally absorbed in boron or other non-fertile absorbers, or would be consumed by the absorption resonances of the fertile materials ^{238}U or ^{232}Th, thereby increasing the fissile production and conversion ratio.

As fuel burnup proceeds the D_2O is replaced by H_2O to maintain reactivity, and the process is repeated for each refueling cycle. Typically, at the beginning of a refueling cycle the reactor coolant would consist of about 75% D_2O. During the cycle the coolant is diluted with normal water, resulting in a concentration of less than 2 to 5% D_2O at the end of the one-year cycle. A facility must be provided to reconcentrate the heavy water. The spectral shift reactor received some attention over a decade ago, but it was not justified economically at that time. Preliminary estimates (S1) indicate significant improvement in conversion ratio over that of any of the light water cycles listed in Table 3.3, even using the lattice of present-day pressurized water reactors. For fuel burnups of 33.4 megawatt day per kg a ^{235}U-Th loading was calculated to operate at an integral conversion ratio of 0.7 and a ^{233}U-Th loading at a conversion ratio as high as 0.87. This indicates the possibility of reducing the lifetime ore requirements well below 2300 metric tons of natural uranium.

Benefits from the higher conversion ratios of spectral shift LWR's must be balanced against increased costs resulting from the increased complexities of using heavy water in LWR's. Facilities must be provided to adjust D_2O concentration in the LWR coolant and to re-enrich the D_2O diluted by H_2O during the fuel cycle. An on-site distillation system for heavy-water enrichment is a possibility.

The presence of concentrated deuterium in the coolant will increase the rate of production of tritium in the coolant. In the pressurized water reactor this increase in tritium production by neutron absorption in deuterium will be offset, in part, by the loss of tritium production from fast-neutron reactions in dissolved boron, since dissolved boron will no longer be needed for reactivity control. A higher concentration of tritium in the coolant may complicate the open-core refueling techniques now used in LWR's, because of the possibility of tritium escape during refueling. It may also require additional controls to minimize the environmental release of tritium via non-condensable off gases during normal operation.

Control of burnup reactivity by spectral shift boiling-water reactors would eliminate the burnable-poison absorbers now incorporated in the fuel rods in these reactors. However, since the burnable absorbers also provide an effective means of adjusting the axial distribution of neutron flux and power density in these reactors, some other technique must be developed for power-density control in a spectral-shift boiling-water reactor. The larger negative void coefficient of reactivity in spectral shift operation would also be a problem.

Nevertheless, the spectral shift concept might be relatively easy to implement in some present PWR's and should be included in further evaluations of alternatives for improved resource utilization. Spectral shift operation with thorium fueling provides the greatest gain in resource utilization, but the improved utilization of uranium fuel with spectral shift operation may become justified when uranium ore prices increase.

There is another way in which thorium may be utilized in a present-day or spectral shift light-water reactor, not merely to extend resources but as a part of an overall approach to international safeguards. As is discussed more completely in Chapter 7, the recycled uranium in a uranium-thorium cycle is diluted with ^{238}U to about 15 to 20% fissile isotopic concentration. This results in somewhat less plutonium production than in a low-enrichment uranium cycle. Such a fuel cycle is completed by storing or reprocessing the discharge fuel to recover the uranium and plutonium.

In summary, the use of thorium in present light water reactors offers a real but marginal advantage for resource extension alone. Larger benefits are possible with redesign of the reactor core or of the moderator-coolant system. Were the fast breeder reactor to be delayed or eliminated altogether, it might be desirable to introduce the use of thorium in LWR's modified for higher conversion ratios, since the overall reduction in uranium ore demand for a larger number of LWR's could be important.

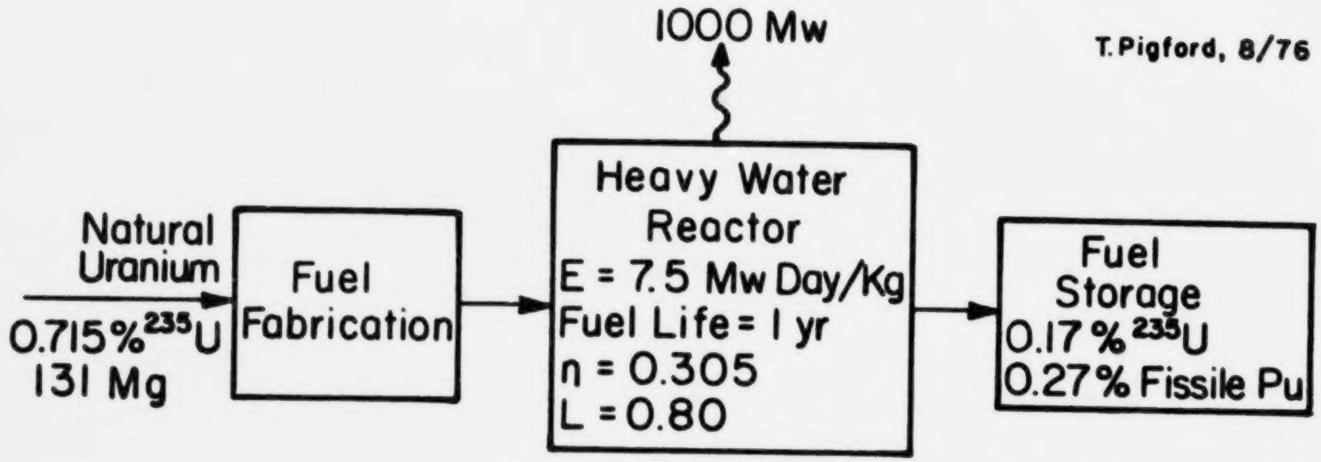
4. Fuel Cycles for Heavy-Water Reactors

4.1 Uranium Fueling as a Reference Case

The Canadian (CANDU) version of the heavy water reactor is being considered as a possible means of better resource utilization in the U.S., if such conservation should become necessary because of delays in the breeder program. The flowsheet for the natural-uranium version of the CANDU reactor (F1, M1) is shown in Figure 4.1. Greater quantities of plutonium are present in the total discharge fuel from this reactor than in the case of a light water reactor of the same power, because of the higher conversion ratio and shorter fuel irradiation exposure of the heavy water reactor. However, the large throughput of uranium results in a relatively low concentration of plutonium in the discharge fuel. Present costs of uranium and of fuel-cycle operations do not now justify reprocessing to recover the plutonium from the fuel discharged from these heavy water reactors, so the discharge fuel is now put into long-term storage. However, future higher costs of uranium ore may ultimately justify reprocessing the fuel to recover and recycle plutonium. As shown in Table 4.1, the ore utilization of the non-recycle CANDU is 25% better than that for the uranium-fueled PWR without recycle.

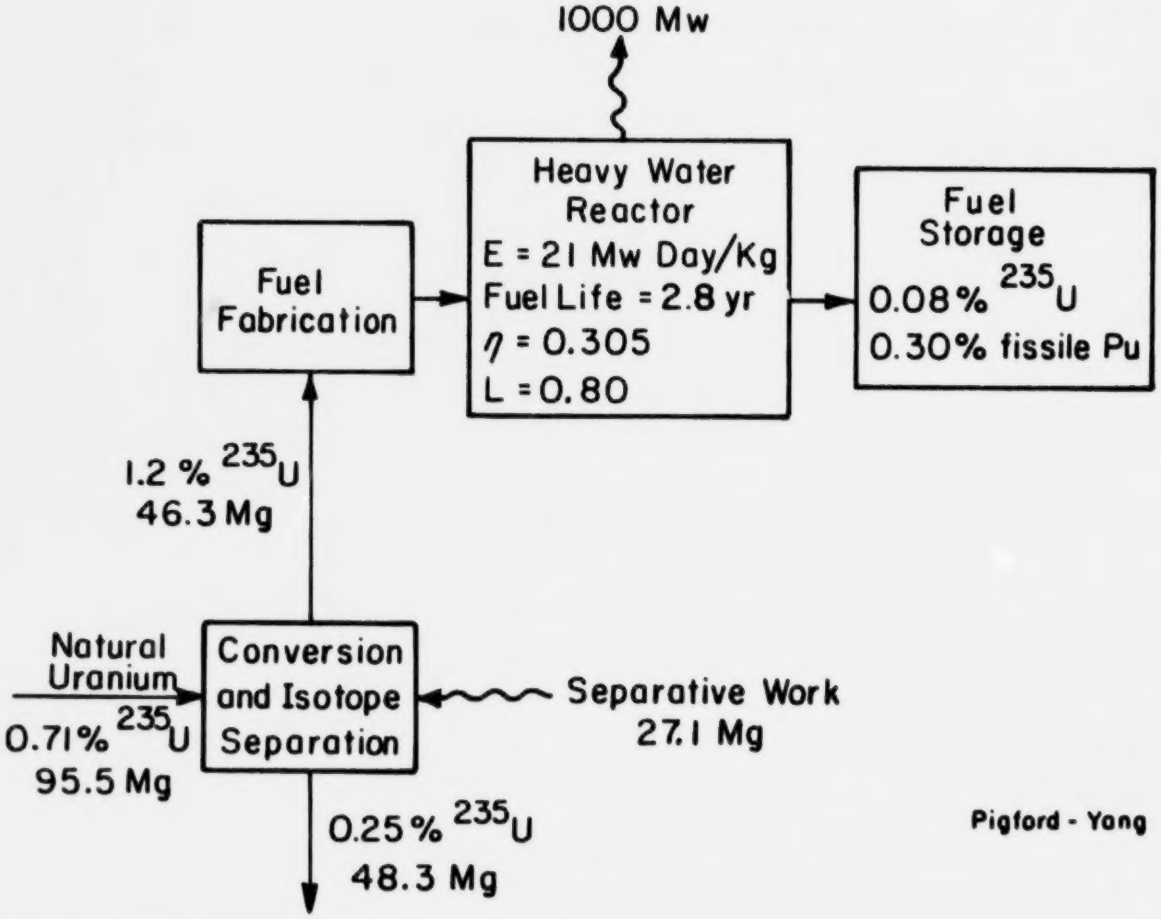
Even without fuel reprocessing, the ore consumption of the CANDU reactor can be reduced by fueling with slightly enriched uranium, as illustrated in Figure 4.2. By increasing the enrichment to 1.2%, the average fuel exposure is increased from 7.5 Mw day/kg to 21 Mw day/kg (B3, T1). Because of the greater burnup, the consumption of natural uranium is only 71% of that of the natural-uranium-fueled CANDU reactor. The concentration of plutonium in the discharge fuel increases, but the total amount of plutonium in the discharge fuel is only 30% of that from the natural-uranium-fueled CANDU reactor.

The calculated operation of the heavy-water reactor with natural uranium and self-generated plutonium recycle (B3) is illustrated in Figure 4.3. Recycling the plutonium makes a significant difference in the fuel burnup, which rises from the low value of 7.5 Mw days/kg for natural uranium to 18 Mw days/kg. The data in this flowsheet were derived from calculations which assumed that the plutonia-urania fuel with the same fuel and cladding dimensions as the present CANDU fuel can operate to the higher burnups without modification. This is an optimistic assumption, since the higher burnups will generate more fission gases. Fission-gas plenums and thicker fuel cladding may be required. As shown in Table 4.1 the lifetime uranium ore requirements for the CANDU reactor with self-generated plutonium recycle are about two-fold less than for the present non-recycle operation with natural uranium. Although for near-term ore costs the burnup per cycle is still too low for reprocessing and recycle to be more economical than the non-reprocessing stowaway cycle, at some future higher price of


1000 Mw
Natural Uranium
0.715% ^{235}U
131 Mg
Fuel Fabrication
Heavy Water Reactor
E = 7.5 Mw Day/Kg
Fuel Life = 1 yr
η = 0.305
L = 0.80
Fuel Storage
0.17% ^{235}U
0.27% Fissile Pu

T. Pigford, 8/76

Fig. 4.1 Annual quantities for natural-U-fueled CANDU reactor (E=fuel exposure, η=overall thermal efficiency, L=capacity factor)


1000 Mw
Fuel Fabrication
Heavy Water Reactor
E = 21 Mw Day/Kg
Fuel Life = 2.8 yr
η = 0.305
L = 0.80
Fuel Storage
0.08% ^{235}U
0.30% fissile Pu
1.2% ^{235}U
46.3 Mg
Natural Uranium
0.71% ^{235}U
95.5 Mg
Conversion and Isotope Separation
Separative Work
27.1 Mg
0.25% ^{235}U
48.3 Mg

Pigford - Yang

Fig. 4.2 Annual quantities for slightly enriched U-fueled CANDU reactor (E=fuel exposure, η=overall thermal efficiency, L=capacity factor)

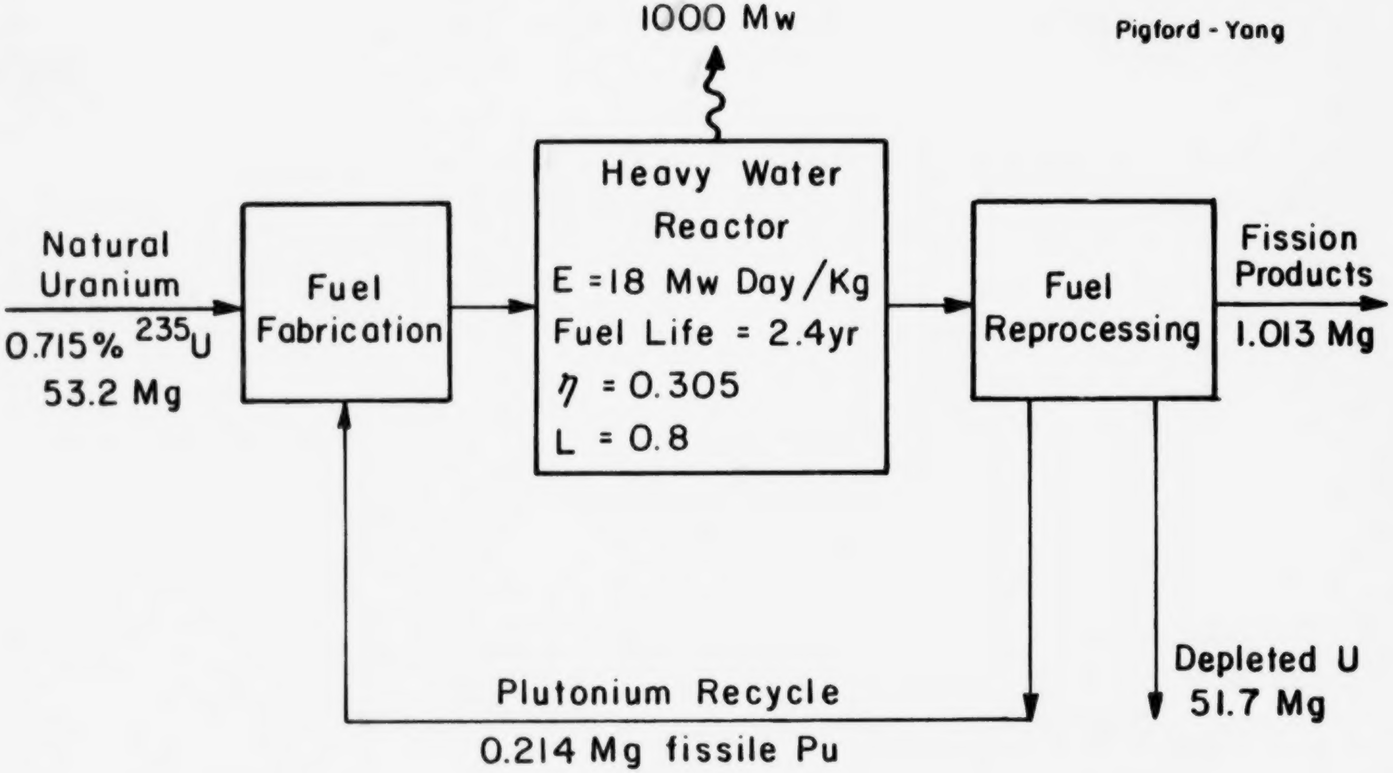

1000 Mw
Pigford - Yang
Natural Uranium
0.715% ^{235}U
53.2 Mg
Fuel Fabrication
Heavy Water Reactor
E = 18 Mw Day/Kg
Fuel Life = 2.4yr
η = 0.305
L = 0.8
Fuel Reprocessing
Fission Products
1.013 Mg
Depleted U
51.7 Mg
Plutonium Recycle
0.214 Mg fissile Pu

Fig. 4.3 Annual quantities for equilibrium U-fueled CANDU reactor, with self generated Pu recycle (E=fuel exposure, η=overall thermal efficiency, L=capacity factor)

TABLE 4.1. 30-Year Lifetime Ore Requirements for Heavy Water Reactors (1000 Mw electrical power, 80% capacity factor)

	Fuel Cycle	Conversion Ratio	Natural Uranium [a] Metric tons (Mg)	Natural Uranium Relative to U-fueled PWR without reprocessing [b]	Thorium [c] Metric tons (Mg)
(a)	Natural U fuel, no recycle	0.75	4060	0.75	
(b)	1.2% ^{235}U in U, no recycle,				
	0.20% depleted U	0.64	2723	0.50	
	0.25% depleted U		2860	0.53	
(c)	Natural U fuel, Pu recycle	0.74	2200	0.41	
(d)	^{235}U-Th fuel, U recycle				
	0.20% depleted U	0.92	1303	0.24	1126
	0.25% depleted U		1442	0.27	
(e)	Pu-Th fuel, U-Pu recycle [d]	0.92	1765	0.33	616

---

[a] Uranium contained in U_3O_8 concentrate. To obtain short tons of U_3O_8 multiply by 1.297.

[b] For 0.25% depleted uranium from isotope separation.

[c] Thorium contained in ThO_2 concentrate. To obtain short tons of ThO_2 multiply by 1.2517.

[d] Includes U-fueled CANDU to supply make-up. Total system power - 1000 Mw.

uranium ore such reprocessing fuel cycles could become economically attractive.

4.2 Thorium-Fueled Heavy-Water Reactors

The same CANDU reactor can also be fueled with thorium and make-up fissile material derived from an external source (B4, C2, K1, T1, T2). Figure 4.4 is the flowsheet for the equilibrium cycle of the CANDU reactor fueled with 93.5% ^{235}U, thorium, and recycled uranium. The fuel burnup has been specified at 27 megawatt days per kilogram of heavy metal, near that typical of light-water fuel. As shown in Table 4.1, uranium-thorium fueling increases the average conversion ratio to 0.92, a result of the greater number of fission neutrons per absorption for the bred and recycled ^{233}U. The uranium ore consumption is 39 to 45% less for this cycle than for uranium fueling with self-generated plutonium recycle. Alternatively, the make-up fissile material for the thorium-fueled CANDU reactor can be plutonium recovered from uranium fuel discharged from a CANDU reactor or a light water reactor. The flowsheet for this cycle at equilibrium, utilizing plutonium produced in a natural uranium CANDU reactor, is shown in Figure 4.5. The lifetime ore requirements for the plutonium-thorium CANDU reactor, shown in Table 4.1, are calculated for 30x0.8 Gw yr of electrical energy from a reactor system consisting of a uranium-fueled CANDU reactor to provide the start-up and make-up plutonium and a plutonium-thorium fueled CANDU to consume the plutonium. The uranium ore required for this system is 20% less than for the CANDU fueled with natural uranium and self-generated plutonium recycle, and it is 22 to 36% greater than for the ^{235}U-Th-fueled CANDU with uranium recycle.

In the present conceptual design of a thorium-fueled CANDU reactor the lattice spacing and specific power have been kept the same as for the natural-uranium CANDU reactor. Because the fuel burnup chosen for these thorium cycles is 3.6 times greater than for present CANDU uranium fuel, the void volume in each fuel rod has been increased by 9% to provide for the accumulation of fission gases (T1). The cladding dimensions have been kept the same, although the higher burnups may require thicker cladding. There are no published data on the performance of CANDU fuel elements at these high burnups.

The lifetime ore requirements for the CANDU with recycle are significantly less than the ore requirements for any of the light water reactor fuel cycles shown in Table 4.1. This is indicative of the overall higher conversion ratio of the heavy water reactor. However, the ore savings and savings in separative work must be balanced against the higher fabrication and reprocessing costs resulting from the lower fuel burnup of the heavy water reactor, differences in construction costs, and the cost of heavy water.

T. Pigford, 1977

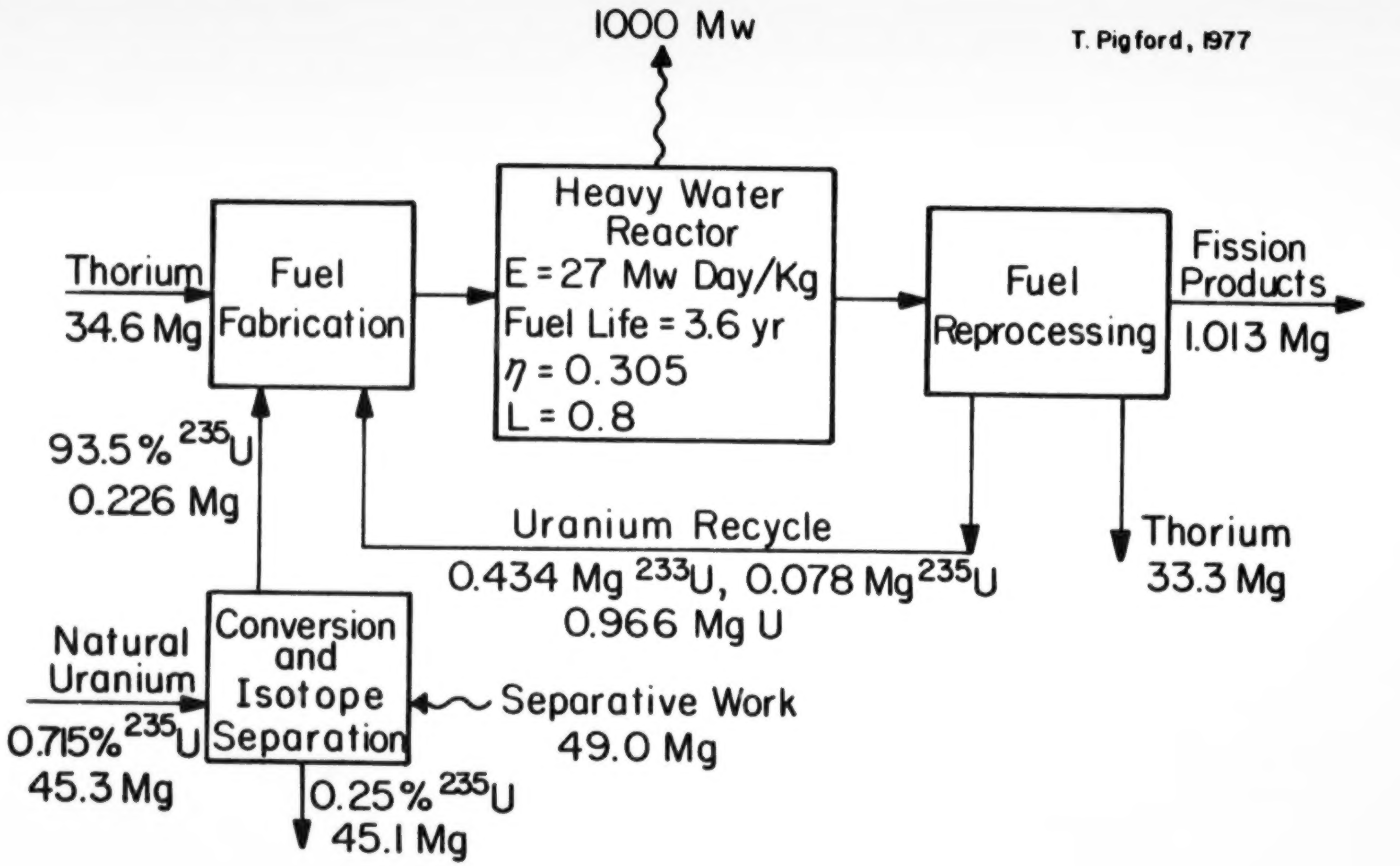
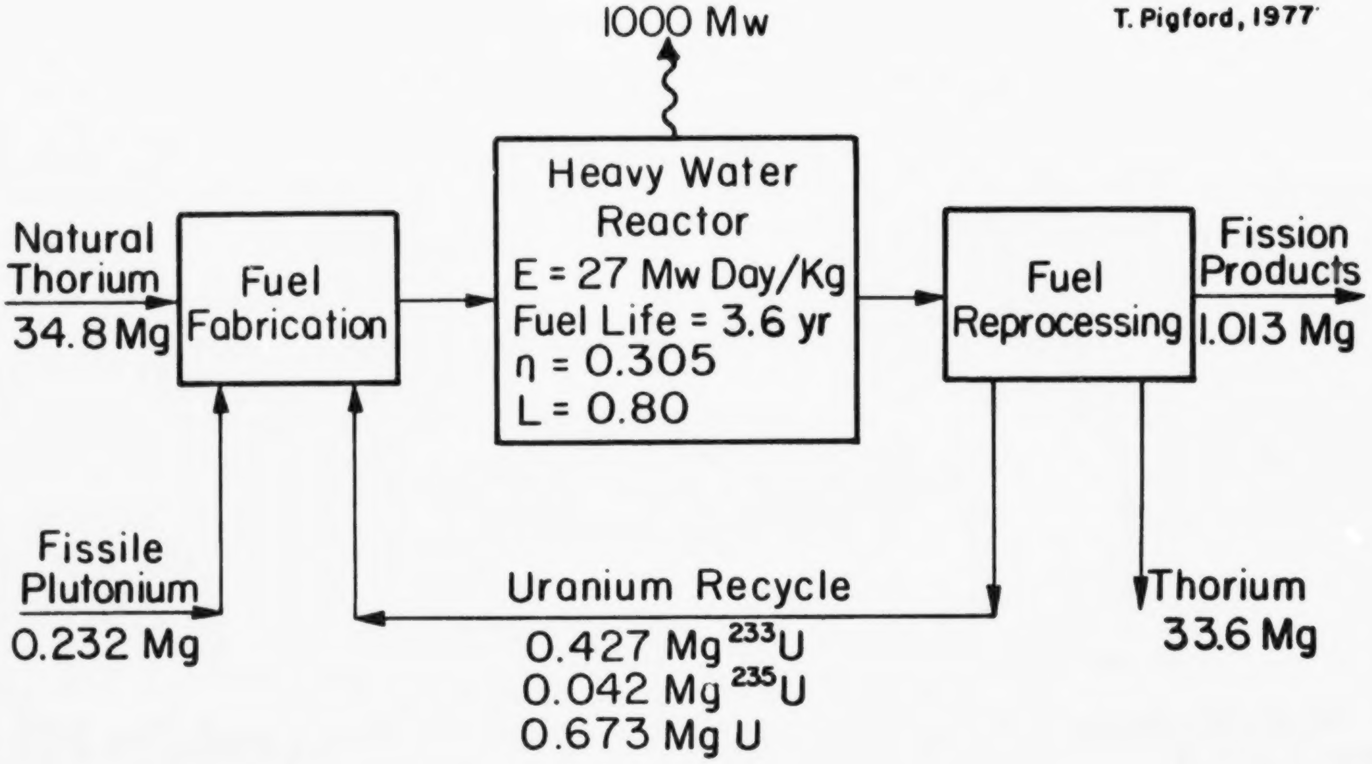

1000 Mw
Thorium
34.6 Mg
Fuel Fabrication
Heavy Water Reactor
E = 27 Mw Day/Kg
Fuel Life = 3.6 yr
η = 0.305
L = 0.8
Fuel Reprocessing
Fission Products
1.013 Mg
93.5 % ^{235}U
0.226 Mg
Uranium Recycle
0.434 Mg ^{233}U, 0.078 Mg ^{235}U
0.966 Mg U
Thorium
33.3 Mg
Natural Uranium
0.715% ^{235}U
45.3 Mg
Conversion and Isotope Separation
Separative Work
49.0 Mg
0.25 % ^{235}U
45.1 Mg

Fig. 4.4 Annual quantities for equilibrium ^{235}U-Th-fueled CANDU reactor, with U recycle (E=fuel exposure, η=overall thermal efficiency, L=capacity factor)


1000 Mw
Heavy Water
Reactor
E = 27 Mw Day/Kg
Fuel Life = 3.6 yr
η = 0.305
L = 0.80
Natural
Thorium
34.8 Mg
Fuel
Fabrication
Fissile
Plutonium
0.232 Mg
Fuel
Reprocessing
Fission
Products
1.013 Mg
Thorium
33.6 Mg
Uranium Recycle
0.427 Mg 233U
0.042 Mg 235U
0.673 Mg U

T. Pigford, 1977

Fig. 4.5 Annual quantities for equilibrium Pu-Th-fueled CANDU reactor, with U recycle (E=fuel exposure, η=overall thermal efficiency, L=capacity factor)

A CANDU-type heavy water reactor can be modified to operate at yet higher conversion ratios, even as a thermal breeder with thorium make-up and uranium recycle (B3, B4, C2, K1, T1, T2). A conversion ratio of unity can theoretically be obtained in the present CANDU lattice if fueled with thorium at low fuel burnup. The conversion ratio can also be increased by increasing the thorium loading, operating at lower specific power, and increasing the calandria lattice spacing. Although the ultimate fuel savings from the higher conversion ratios are ultimately very large, they cannot be realized at the beginning of the reactor lifetime. A higher initial fissile loading is required, and the smaller cumulative ore requirements are realized only after many years of operation.

The startup of the CANDU thorium breeder has been studied by Banerjee, et al. (B6, B11). Prebreeding is initiated by fueling the CANDU reactor with thorium and with plutonium recovered from irradiated CANDU uranium. The discharged thorium-plutonium fuel is reprocessed, and recovered plutonium and bred uranium are recycled with additional make-up or recycled thorium. The cycle is subsequently repeated. Additional plutonium is added to each recycle loading to maintain reactivity, but the necessary amount of plutonium decreases with each subsequent cycle, until the reactor finally becomes self-sustaining on thorium and recycled bred uranium.

For reactors with characteristics of the present CANDU reactors, self-sustaining thorium breeding occurs at fuel exposures of 12.1 Mw day/kg. The breeding ratio is unity, so all subsequent breeders must be started up by the same process of utilizing plutonium recovered from irradiated uranium fuel, or, alternatively, obtaining enriched uranium from isotope separation. Based upon startup with CANDU plutonium, and using the cycle-by-cycle data of Banerjee and Barclay (B11) for Pu-Th fueling, with Pu-U recycle, the total plutonium required to achieve self-sustaining breeder is 4410 kg for a 1000 Mw CANDU. The total amount of natural uranium to produce this amount of plutonium in a CANDU is 1650 Mg.

The time-dependent demand for U uranium ore for startup of a 1000 Mw CANDU thorium breeder is shown in Figure 4.6. One approach is to obtain an initial amount of plutonium of 1962 kg, sufficient for a Pu-Th initial loading. To produce this plutonium, 734 Mg of natural uranium would be required to fuel a natural-uranium CANDU reactor. This corresponds to the initial ore requirement of curve 1 of Figure 4.6. In subsequent cycles additional plutonium is required, so additional uranium must be irradiated in separate CANDU reactors to produce that plutonium. Assuming two years for reprocessing and fabrication of recycled fuel, the reactor becomes self-sustaining in 10.8 years. This scenario is realistic when there exists a stored inventory of irradiated uranium fuel to be reprocessed for the startup plutonium inventory, or when there are existing uranium-fueled CANDU reactors to produce the plutonium needed for thorium breeder startup.

If an inventory of irradiated uranium is not available, or if uranium-fueled CANDU reactors are not available, the CANDU reactor destined to become a thorium breeder can be operated initially on uranium fueling for 12.6 years, at which time sufficient plutonium has been produced to convert the entire reactor to Pu-Th fueling. Self-sustaining breeding occurs after 23.4 years.

The demand for uranium ore for startup of CANDU thorium breeders can be spread over a longer period of time by starting the CANDU with uranium fueling

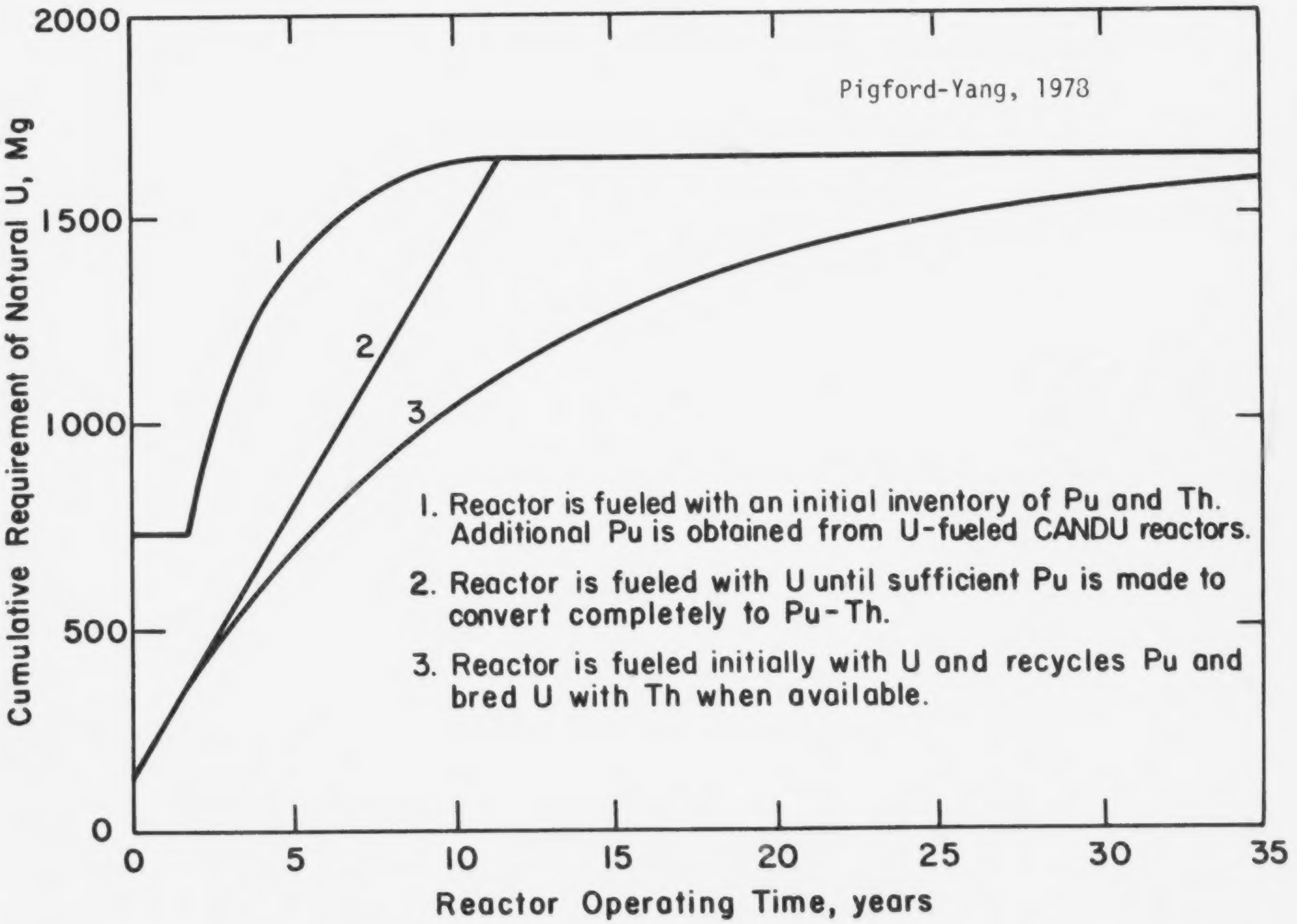


Figure 4.6 Cumulative Requirement of Natural Uranium for the Pu-Topped, Thorium-Fueled Self-Sufficient CANDU reactor (1000 Mwe, fuel exposure at equilibrium = 12.1 Mwday/kg)

and recycling the self-generated plutonium and bred uranium as Pu-Th-U fuel, as it becomes available. In each subsequent cycle some of the reactor pressure tubes previously fueled with natural uranium are converted to thorium fueling. Thorium fuel elements progress through the calandria tube more slowly than do the uranium fuel elements, because of the higher burnup for thorium breeding. After 51 years the reactor becomes a self-sustaining thorium breeder, with 99% of the pressure tubes operating on thorium fuel.

The total ore requirement for breeder startup is the same for each of the three different approaches considered above.

As seen above, to produce the total plutonium required to start the thorium CANDU breeder requires the operation of 1000 Mwe uranium-fueled CANDU reactor for 12.2 years on natural-uranium fuel, consuming 1650 Mg of natural uranium. If, instead, the plutonium recovered from the discharge uranium fuel were recycled as mixed-oxide fuel with natural uranium, the ore consumption of this original CANDU reactor for 12.2 years could have been reduced to 895 Mg of natural uranium, a saving of 800 Mg. Therefore, the uranium ore directly attributable to breeder start-up is 800 Mg. This means that 800 Mg of additional natural uranium would be required in the original CANDU reactor if the plutonium in the discharge fuel is to be accumulated for later breeder startup, instead of utilizing the plutonium, when available, in self-generated recycle to reduce the consumption of natural uranium.

5. High-Temperature Gas-Cooled Reactor

5.1 Reactor Characteristics

The high-temperature gas cooled reactor (HTGR) moderated with graphite and cooled with helium, is undergoing demonstration tests as an alternative nuclear power plant of the future. As shown in the overall material flowsheet of Figure 5.1 for the HTGR reference design, the reactor is fueled with thorium, make-up ^{235}U, and recycled uranium. The overall flowsheet of Figure 5.1 is a simplified composite of the more detailed flowsheet of Figure 5.2, which shows the several different and segregated fuel streams, scrap recycle (J1), and process losses.

The fuel consists of coated particles of uranium and thorium embedded in a prismatic graphite matrix. Helium coolant flows through holes in the graphite. The fuel-moderator prisms, of hexagonal cross section, are stacked to form the core structure. The graphite matrix provides a means of obtaining very high fuel burnup without loss of mechanical integrity. The current design burnup is 94.3 megawatt days per kilogram of uranium and thorium, which is about three times that experienced in light water reactors. A commercial prototype is now operating at Fort St. Vrain in Colorado. However, the U.S. manufacturer, General Atomic, has recently withdrawn its earlier sales of full-scale commercial plants. Development of HTGR fuel reprocessing and refabrication technology continues under DOE sponsorship. It is uncertain when and if this reactor will return to the commerical U.S. market.

Although the overall HTGR fuel cycle appears similar to the ^{235}U-thorium fuel cycles for the light-water reactor (Figure 3.5) and for the heavy-water reactor (Figure 4.4), the HTGR flowsheet differs in detail because of the plans to segregate the various fissile and fertile fuel streams for the purpose of reprocessing and recycle. This is shown in the more detailed flowsheet of Figure 5.2. The reprocessing flowsheet further differs from that for the thoria fuels of the light-water and heavy-water reactors because of the large quantity of graphite in the HTGR fuel matrix. In HTGR fuel reprocessing this graphite is burned to expose the fuel particles for acid dissolution.

The discharge fuel is processed to recover the uranium remaining from the initial make-up ^{235}U, which is then recycled for one more pass through the reactor. Also recovered for recycling is the uranium, largely fissile ^{233}U, formed by neutron-capture reactions in thorium.

Corresponding material quantities for the near-equilibrium fuel cycle, derived from data for the ninth reload (H4), are given in Tables 5.1, 5.2, 5.3, and 5.4. The actinide quantities in the composite of the three discharge-fuel streams are given in Table 5.5.

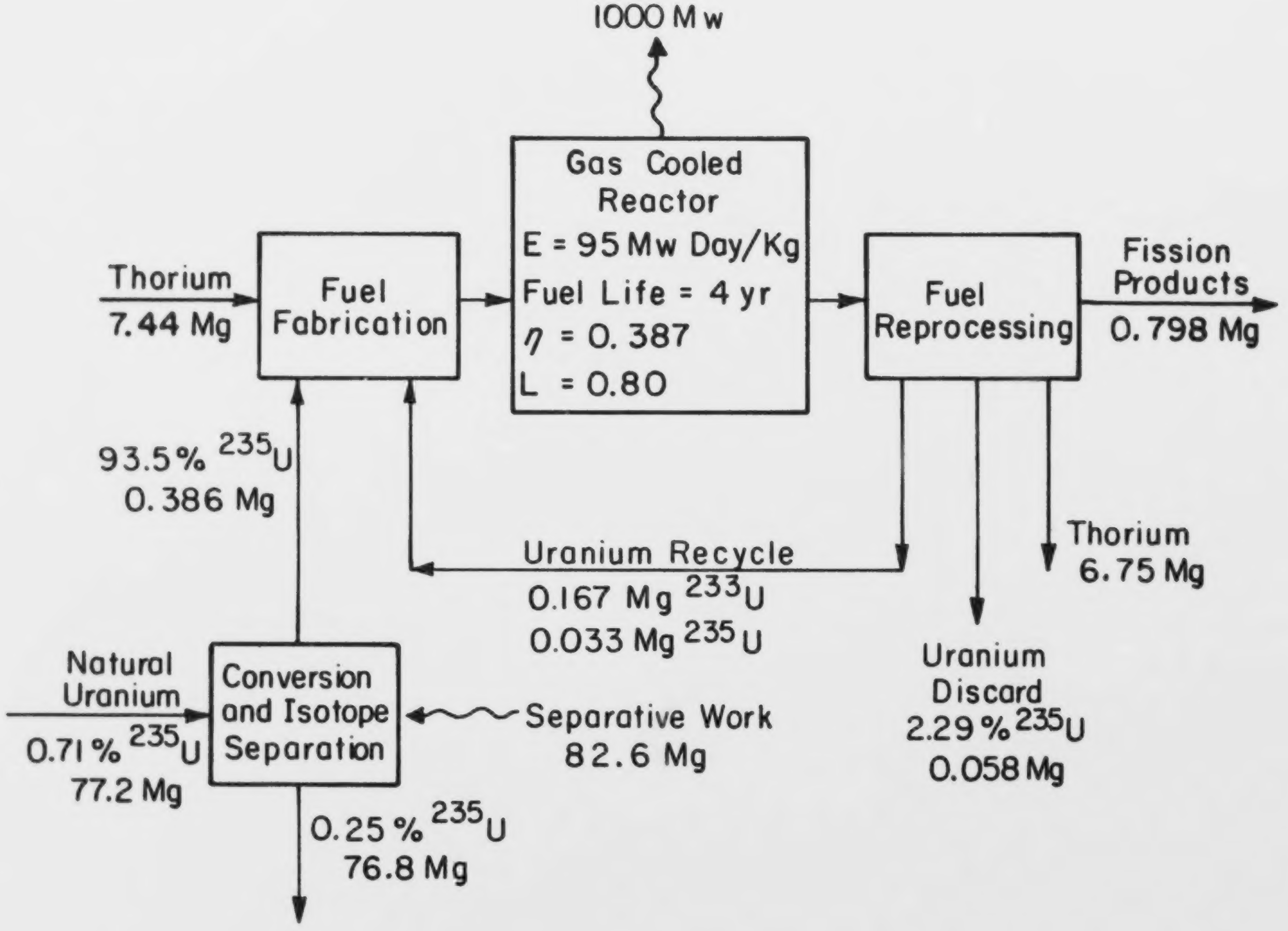

1000 Mw
Gas Cooled Reactor
E = 95 Mw Day/Kg
Fuel Life = 4 yr
η = 0.387
L = 0.80
Thorium
7.44 Mg
Fuel Fabrication
Fuel Reprocessing
Fission Products
0.798 Mg
93.5% ^{235}U
0.386 Mg
Uranium Recycle
0.167 Mg ^{233}U
0.033 Mg ^{235}U
Thorium
6.75 Mg
Uranium Discard
2.29% ^{235}U
0.058 Mg
Natural Uranium
0.71% ^{235}U
77.2 Mg
Conversion and Isotope Separation
Separative Work
82.6 Mg
0.25% ^{235}U
76.8 Mg

Fig. 5.1 Annual quantities for the near equilibrium ^{235}U-Th-fueled HTGR, with U recycle (E=fuel exposure, η=overall thermal efficiency, L=capacity factor)

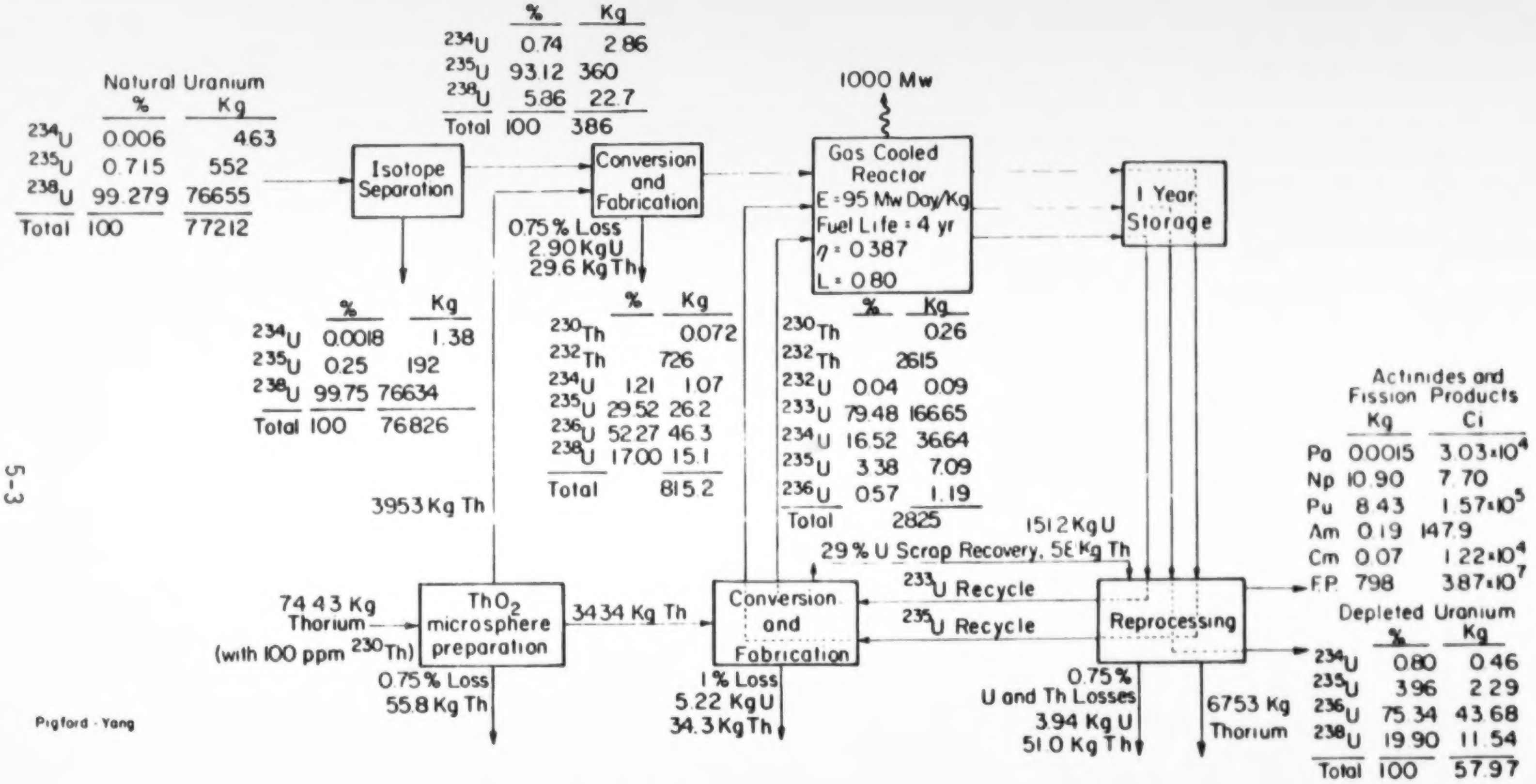

Natural Uranium
% Kg
234U 0.006 463
235U 0.715 552
238U 99.279 76655
Total 100 77212
Isotope Separation
% Kg
234U 0.0018 1.38
235U 0.25 192
238U 99.75 76634
Total 100 76826
% Kg
234U 0.74 2.86
235U 93.12 360
238U 5.86 22.7
Total 100 386
Conversion and Fabrication
0.75% Loss
2.90 Kg U
29.6 Kg Th
% Kg
230Th 0.072
232Th 726
234U 1.21 1.07
235U 29.52 26.2
236U 52.27 46.3
238U 17.00 15.1
Total 815.2
3953 Kg Th
74.43 Kg Thorium (with 100 ppm 230Th)
ThO2 microsphere preparation
0.75% Loss
55.8 Kg Th
3434 Kg Th
1000 Mw
Gas Cooled Reactor
E = 95 Mw Day/Kg
Fuel Life = 4 yr
η = 0.387
L = 0.80
% Kg
230Th 0.26
232Th 2615
232U 0.04 0.09
233U 79.48 166.65
234U 16.52 36.64
235U 3.38 7.09
236U 0.57 1.19
Total 2825
1512 Kg U
29% U Scrap Recovery, 58 Kg Th
233U Recycle
235U Recycle
Conversion and Fabrication
1% Loss
5.22 Kg U
34.3 Kg Th
1 Year Storage
Reprocessing
0.75% U and Th Losses
3.94 Kg U
51.0 Kg Th
6753 Kg Thorium
Actinides and Fission Products
Kg Ci
Pa 0.0015 3.03×10^4
Np 10.90 7.70
Pu 8.43 1.57×10^5
Am 0.19 147.9
Cm 0.07 1.22×10^4
F.P. 798 3.87×10^7
Depleted Uranium
% Kg
234U 0.80 0.46
235U 3.96 2.29
236U 75.34 43.68
238U 19.90 11.54
Total 100 57.97
Pigford - Yang

Fig. 5.2 Detailed annual mass flow sheet for the near-equilibrium ^{235}U-Th-fueled HTGR, with U recycle (E=fuel exposure, η=overall thermal efficiency, L=capacity factor)

TABLE 5.1

Actinides in the Fuel Charged to the U-Th Fueled HTGR

(1000 Mwe, high-temperature gas-cooled reactor with uranium recycle[a])

1. ^{235}U-Th make-up fuel[b]

Radionuclide		kg/yr	Ci/yr	Weight %
Thorium	230	$3.89x10^{-01}$	7.56	0.01
	232	$3.92x10^{+03}$	$4.29x10^{-01}$	99.99
Total		$3.92x10^{+03}$	7.99	100.00
Uranium	234	2.84	$6.66x10^{+03}$	0.74
	235	$3.58x10^{+02}$	$7.67x10^{-01}$	93.12
	236	1.09	$6.82x10^{+01}$	0.28
	238	$2.25x10^{+01}$	$7.50x10^{-03}$	5.86
Total		$3.84x10^{+02}$	$6.73x10^{+03}$	100.00

2. Once recycled ^{235}U-Th fuel[b], [c]

Radionuclide		kg/yr	Ci/yr	Weight %
Thorium	230	$7.20x10^{-02}$	1.40	0.01
	232	$7.26x10^{+02}$	$7.94x10^{-02}$	99.99
Total		$7.26x10^{+02}$	1.48	100.00
Uranium	234	1.07	$6.66x10^{+03}$	1.21
	235	$2.62x10^{+01}$	$5.61x10^{-02}$	29.52
	236	$4.63x10^{+01}$	2.94	52.27
	238	$1.51x10^{+01}$	$5.02x10^{-03}$	17.00
Total		$8.87x10^{+01}$	$6.66x10^{+03}$	100.00

3. Recycled ^{233}U-Th fuel[b], [c]

Radionuclide		kg/yr	Ci/yr	Weight %
Thorium	230	$2.59x10^{-01}$	5.04	0.01
	232	$2.62x10^{+03}$	$2.86x10^{-01}$	99.99
Total		$2.62x10^{+03}$	5.33	100.00
Uranium	232	$9.00x10^{-02}$	$1.93x10^{+03}$	0.04
	233	$1.67x10^{+02}$	$1.58x10^{+03}$	79.49
	234	$3.46x10^{+01}$	$2.14x10^{+02}$	16.52
	235	7.09	2.31	3.38
	236	1.19	$6.82x10^{+01}$	0.57
Total		$2.10x10^{+02}$	$3.79x10^{+03}$	100.00

[a] 95 Mw-day/kg HM, 38.7% thermal efficiency, 80% capacity factor, near-equilibrium fuel cycle.

[b] Natural thorium is assumed to contain 100 ppm ^{230}Th.

[c] 1 year cooling of discharged fuel before reprocessing, 0.75% loss in reprocessing, 1% loss in fabrication. Thorium is added to the recycled uranium to dilute the recycle-fuel to proper enrichment.

TABLE 5.2

Actinides in Discharged ^{235}U-Th First Cycle Make-Up HTGR Fuel
(1000 Mwe U-Th fueled high-temperature gas-cooled reactor[a])

Radionuclide		Fissile Fuel		Fertile Fuel	
		kg/yr	Ci/yr	kg/yr	Ci/yr
Thorium[b]	228	1.26×10^{-7}	1.04×10^{-1}	7.72×10^{-4}	6.34×10^{2}
	229	1.39×10^{-8}	2.96×10^{-6}	8.37×10^{-4}	1.78×10^{-1}
	230	1.77×10^{-5}	3.44×10^{-4}	1.43×10^{-1}	2.77
	231	1.22×10^{-10}	6.49×10^{-2}	1.62×10^{-11}	8.62×10^{-3}
	232	4.55×10^{-6}	4.98×10^{-10}	3.64×10^{3}	3.98×10^{-1}
	234	1.22×10^{-12}	2.82×10^{-5}	2.75×10^{-8}	6.36×10^{-1}
Total		2.24×10^{-5}	$\alpha = 1.04\times10^{-1}$ $\beta = 6.49\times10^{-2}$	3.64×10^{3}	$\alpha = 6.37\times10^{2}$ $\beta = 6.45\times10^{-1}$
Protactinium	233	1.04×10^{-12}	2.16×10^{-5}	7.88×10^{-4}	1.64×10^{4}
	234	1.42×10^{-17}	2.82×10^{-8}	3.21×10^{-13}	6.36×10^{-4}
Total		1.04×10^{-12}	$\beta = 2.16\times10^{-5}$	7.88×10^{-4}	$\beta = 1.64\times10^{4}$
Uranium[c]	232	9.43×10^{-6}	2.02×10^{-1}	5.56×10^{-2}	1.19×10^{3}
	233	1.49×10^{-6}	1.41×10^{-5}	9.72×10^{1}	9.21×10^{2}
	234	1.23	7.61	1.94×10^{1}	1.20×10^{2}
	235	2.98×10^{1}	6.39×10^{-2}	3.95	8.47×10^{-3}
	236	5.28×10^{1}	3.35	$6.26-10^{-1}$	3.97×10^{-2}
	237	9.54×10^{-9}	7.79×10^{-1}	4.71×10^{-12}	3.85×10^{-4}
	238	1.72×10^{1}	5.73×10^{-3}	2.72×10^{-5}	9.03×10^{-9}
Total		1.01×10^{2}	$\alpha = 1.12\times10^{1}$ $\beta = 7.79\times10^{-1}$	1.21×10^{2}	$\alpha = 2.23\times10^{3}$ $\beta = 3.85\times10^{-4}$
Neptunium	237	6.46	4.56	4.04×10^{-2}	2.85×10^{-2}
Total		6.46	$\alpha = 4.56$	4.04×10^{-2}	$\alpha = 2.95\times10^{-2}$
Plutonium	236	2.52×10^{-6}	1.34	1.02×10^{-12}	5.43×10^{-7}
	238	3.14	5.49×10^{4}	9.83×10^{-3}	1.72×10^{2}
	239	6.82×10^{-1}	4.13×10^{1}	7.67×10^{-4}	4.70×10^{-2}
	240	3.17×10^{-1}	7.18×10^{1}	2.40×10^{-4}	5.44×10^{-2}
	241	2.96×10^{-1}	3.33×10^{4}	1.46×10^{-4}	1.65×10^{1}
	242	3.11×10^{-1}	1.21	5.40×10^{-5}	2.11×10^{-4}
Total		4.75	$\alpha = 5.50\times10^{4}$ $\beta = 3.30\times10^{4}$	1.10×10^{-2}	$\alpha = 1.72\times10^{2}$ $\beta = 1.65\times10^{1}$
Americium	241	2.03×10^{-2}	6.58×10^{1}	8.75×10^{-6}	2.84×10^{-2}
	242m	1.72×10^{-4}	1.67	2.78×10^{-7}	2.70×10^{-3}
	243	9.00×10^{-2}	1.67×10^{1}	2.28×10^{-5}	4.22×10^{-3}
Total		1.10×10^{-1}	$\alpha = 8.25\times10^{1}$ $\beta = 1.67$	3.18×10^{-5}	$\alpha = 3.26\times10^{-2}$ $\beta = 2.70\times10^{-3}$
Curium	242	1.13×10^{-3}	3.74×10^{3}	1.82×10^{-7}	6.02×10^{-1}
	243	7.45×10^{-5}	3.43	5.56×10^{-9}	2.56×10^{-4}
	244	4.00×10^{-2}	3.33×10^{3}	4.50×10^{-5}	3.75
	245	1.68×10^{-4}	2.64×10^{-2}	2.52×10^{-7}	3.95×10^{-5}
Total		4.14×10^{-2}	$\alpha = 7.07\times10^{3}$	4.54×10^{-5}	$\alpha = 4.35$
TOTAL ACTINIDES		1.12×10^{2}	$\alpha = 6.22\times10^{4}$ $\beta = 3.33\times10^{4}$	3.76×10^{3}	$\alpha = 3.04\times10^{3}$ $\beta = 1.64\times10^{4}$

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[a] 95 Mw-day/kg HM, 38.7% thermal efficiency, 80% capacity factor, 1 year after discharge, near-equilibrium fuel cycle.

[b] Natural thorium is assumed to contain 100 ppm ^{230}Th. Discharged thorium is not recycled.

[c] Initial make-up uranium is 93.1% ^{235}U. Recovered uranium from fissile fuel is recycled as second-cycle uranium-235 fuel.

TABLE 5.3

Actinides in Discharged ^{235}U-Th Second-Cycle HTGR Fuel

(1000 Mwe U-Th fueled high-temperature gas-cooled reactor[a])

Radionuclide		Fissile Fuel		Fertile Fuel	
		kg/yr	Ci/yr	kg/yr	Ci/yr
Thorium[b]	228	$4.80x10^{-8}$	$3.94x10^{-2}$	$1.43x10^{-4}$	$1.17x10^{2}$
	229	$5.29x10^{-9}$	$1.13x10^{-6}$	$1.55x10^{-4}$	$3.30x10^{-2}$
	230	$6.85x10^{-6}$	$1.33x10^{-4}$	$2.64x10^{-2}$	$5.13x10^{-1}$
	231	$9.39x10^{-12}$	$5.00x10^{-3}$	$3.00x10^{-12}$	$1.60x10^{-3}$
	232	$7.47x10^{-6}$	$8.17x10^{-10}$	$6.75x10^{2}$	$7.38x10^{-2}$
	234	$8.42x10^{-13}$	$1.95x10^{-5}$	$5.09x10^{-9}$	$1.18x10^{-1}$
Total		$1.44x10^{-5}$	$\alpha= 3.95x10^{-2}$	$6.75x10^{2}$	$\alpha= 1.18x10^{2}$
			$\beta= 5.02x10^{-3}$		$\beta= 1.20x10^{-1}$
Protactinium	233	$1.92x10^{-12}$	$3.98x10^{-5}$	$1.46x10^{-4}$	$3.03x10^{3}$
	234	$9.83x10^{-18}$	$1.95x10^{-8}$	$5.94x10^{-14}$	$1.18x10^{-4}$
Total		$1.92x10^{-12}$	$\beta= 3.98x10^{-5}$	$1.46x10^{-4}$	$\beta= 3.03x10^{3}$
Uranium[c]	232	$3.72x10^{-6}$	$7.97x10^{-2}$	$1.03x10^{-2}$	$2.21x10^{2}$
	233	$7.35x10^{-7}$	$6.97x10^{-6}$	$1.80x10^{1}$	$1.71x10^{2}$
	234	$4.64x10^{-1}$	2.87	3.59	$2.22x10^{1}$
	235	2.29	$4.91x10^{-3}$	$7.32x10^{-1}$	$1.57x10^{-3}$
	236	$4.37x10^{1}$	2.77	$1.16x10^{-1}$	$7.36x10^{-3}$
	237	$6.90x10^{-9}$	$5.63x10^{-1}$	$8.73x10^{-13}$	$7.13x10^{-5}$
	238	$1.15x10^{1}$	$3.83x10^{-3}$	$5.02x10^{-6}$	$1.67x10^{-9}$
Total		$5.80x10^{1}$	$\alpha= 5.73$	$2.24x10^{1}$	$\alpha= 4.14x10^{2}$
			$\beta= 5.63x10^{-1}$		$\beta= 7.13x10^{-5}$
Neptunium	237	3.79	2.67	$7.49x10^{-3}$	$5.28x10^{-3}$
Total		3.79	$\alpha= 2.67$	$7.49x10^{-3}$	$\alpha= 5.28x10^{-3}$
Plutonium	236	$1.82x10^{-6}$	$9.68x10^{-1}$	$1.89x10^{-13}$	$1.01x10^{-7}$
	238	2.21	$3.86x10^{4}$	$1.82x10^{-3}$	$3.18x10^{1}$
	239	$4.75x10^{-1}$	$2.91x10^{1}$	$1.42x10^{-4}$	$8.17x10^{-3}$
	240	$2.29x10^{-1}$	$5.19x10^{1}$	$4.45x10^{-5}$	$1.01x10^{-2}$
	241	$2.14x10^{-1}$	$2.41x10^{4}$	$2.71x10^{-5}$	3.05
	242	$2.25x10^{-1}$	$8.78x10^{-1}$	$1.00x10^{-5}$	$3.90x10^{-5}$
Total		3.35	$\alpha= 3.87x10^{4}$	$2.04x10^{-3}$	$\alpha= 3.18x10^{1}$
			$\beta= 2.41x10^{4}$		$\beta= 3.05$
Americium	241	$1.49x10^{-2}$	$4.83x10^{1}$	$1.62x10^{-6}$	$5.25x10^{-3}$
	242m	$1.26x10^{-4}$	1.23	$5.14x10^{-8}$	$5.00x10^{-4}$
	243	$6.40x10^{-2}$	$1.19x10^{1}$	$4.22x10^{-6}$	$7.81x10^{-4}$
Total		$7.90x10^{-2}$	$\alpha= 6.02x10^{1}$	$5.89x10^{-6}$	$\alpha= 6.03x10^{-3}$
			$\beta= 1.23$		$\beta= 5.00x10^{-4}$
Curium	242	$8.20x10^{-4}$	$2.71x10^{3}$	$3.37x10^{-8}$	$1.12x10^{-1}$
	243	$5.34x10^{-5}$	2.46	$1.03x10^{-9}$	$4.74x10^{-5}$
	244	$2.80x10^{-2}$	$2.33x10^{3}$	$8.33x10^{-6}$	$6.94x10^{-1}$
	245	$1.18x10^{-4}$	$1.85x10^{-2}$	$4.66x10^{-8}$	$7.32x10^{-6}$
Total		$2.90x10^{-2}$	$\alpha= 5.04x10^{3}$	$8.41x10^{-6}$	$\alpha= 8.06x10^{-1}$
TOTAL ACTINIDES		$6.52x10^{1}$	$\alpha= 4.38x10^{4}$	$6.97x10^{2}$	$\alpha= 5.65x10^{2}$
			$\beta= 2.41x10^{4}$		$\beta= 3.03x10^{3}$

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[a] 95 Mw-day/kg HM, 38.7% thermal efficiency, 80% capacity factor, 1 year after discharge, near equilibrium fuel cycle.

[b] Natural thorium is assumed to contain 100 ppm ^{230}Th. Discharged thorium is not recycled.

[c] Uranium in fuel charged is recovered from discharged first-cycle make-up ^{235}U-Th fissile fuel.

TABLE 5.4

Actinides in Discharged ^{233}U-Th Recycled HTGR Fuel

(1000 Mwe U-Th fueled high-temperature gas-cooled reactor[a])

Radionuclide	Fissile Fuel		Fertile Fuel	
	kg/yr	Ci/yr	kg/yr	Ci/yr
Thorium[b] 228	1.30×10^{-3}	1.07×10^{3}	5.15×10^{-4}	4.23×10^{2}
229	1.82×10^{-3}	3.87×10^{-1}	5.58×10^{-4}	1.19×10^{-1}
230	7.43×10^{-3}	1.44×10^{-1}	9.50×10^{-2}	1.85
231	3.96×10^{-11}	2.11×10^{-2}	1.08×10^{-11}	5.75×10^{-3}
232	2.70×10^{-5}	2.95×10^{-9}	2.43×10^{3}	2.66×10^{-1}
234	2.28×10^{-16}	5.28×10^{-9}	1.83×10^{-8}	4.24×10^{-1}
Total	1.06×10^{-2}	α= 1.07×10^{3} β 2.11×10^{-2}	2.43×10^{3}	α= 1.50×10^{3} β= 4.30×10^{-1}
Protactinium 233	5.49×10^{-12}	1.14×10^{-4}	5.26×10^{-4}	1.09×10^{4}
234	2.66×10^{-21}	5.28×10^{-12}	2.14×10^{-13}	4.24×10^{-3}
Total	5.49×10^{-12}	β= 1.14×10^{-4}	5.26×10^{-4}	β= 1.09×10^{4}
Uranium[c] 232	3.62×10^{-2}	7.75×10^{2}	3.71×10^{-2}	7.94×10^{2}
233	9.84	9.33×10^{1}	6.48×10^{1}	6.14×10^{2}
234	2.44×10^{1}	1.51×10^{2}	1.29×10^{1}	8.00×10^{1}
235	9.65	2.07×10^{-2}	2.64	5.65×10^{-3}
236	4.92	3.12×10^{-1}	4.18×10^{-1}	2.65×10^{-2}
237	2.50×10^{-10}	2.04×10^{-2}	3.14×10^{-12}	2.57×10^{-4}
238	5.02×10^{-4}	1.67×10^{-7}	1.81×10^{-5}	6.02×10^{-9}
Total	4.88×10^{1}	α= 1.02×10^{3} β= 2.04×10^{-2}	8.08×10^{1}	α= 1.49×10^{3} β= 2.57×10^{-4}
Neptunium 237	5.95×10^{-1}	4.20×10^{-1}	2.70×10^{-2}	1.90×10^{-2}
Total	5.95×10^{-1}	α= 4.20×10^{-1}	2.70×10^{-2}	α= 1.90×10^{-2}
Plutonium 236	1.33×10^{-11}	7.07×10^{-6}	6.80×10^{-13}	3.62×10^{-7}
238	2.66×10^{-1}	4.65×10^{3}	6.55×10^{-3}	1.15×10^{2}
239	2.51×10^{-2}	1.54	5.11×10^{-4}	3.14×10^{-2}
240	1.01×10^{-2}	2.29	1.60×10^{-4}	3.63×10^{-2}
241	7.75×10^{-3}	8.71×10^{2}	9.76×10^{-5}	1.10×10^{1}
242	4.04×10^{-3}	1.58×10^{-2}	3.60×10^{-5}	1.40×10^{-4}
Total	3.13×10^{-1}	α= 4.65×10^{3} β= 8.71×10^{2}	7.35×10^{-3}	α= 1.15×10^{2} β= 1.10×10^{1}
Americium 241	5.00×10^{-4}	1.62	5.83×10^{-6}	1.89×10^{-2}
242m	3.23×10^{-6}	3.14×10^{-2}	1.85×10^{-7}	1.80×10^{-3}
243	6.72×10^{-4}	1.24×10^{-1}	1.52×10^{-5}	2.81×10^{-3}
Total	1.18×10^{-3}	α= 1.74 β= 3.14×10^{-2}	2.12×10^{-5}	α= 2.17×10^{-2} β= 1.80×10^{-3}
Curium 242	1.55×10^{-5}	5.13×10^{1}	1.21×10^{-7}	4.02×10^{-1}
243	6.52×10^{-7}	3.00×10^{-2}	3.71×10^{-9}	1.71×10^{-4}
244	2.13×10^{-4}	1.77×10^{1}	3.00×10^{-5}	2.50
245	8.47×10^{-7}	1.33×10^{-4}	1.68×10^{-7}	2.63×10^{-5}
Total	2.30×10^{-4}	α= 6.90×10^{1}	3.03×10^{-5}	2.90
TOTAL ACTINIDES	4.97×10^{1}	α= 6.81×10^{3} β= 8.71×10^{2}	2.51×10^{3}	α= 3.11×10^{3} β= 1.90×10^{4}

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[a] 95 Mw-day/kg HM, 38.7% thermal efficiency, 80% capacity factor, 1 year after discharge, near-equilibrium fuel cycle.

[b] Natural thorium is assumed to contain 100 ppm ^{230}Th. Discharged thorium is not recycled.

[c] Uranium in fuel charged is recovered from discharged fertile fuel and previous cycle discharged ^{235}U-Th fissile fuel

TABLE 5.5

Actinides in Discharge Thorium Fuel

(1000 Mwe U-Th fueled high-temperature gas-cooled HTGR reactor a/)

Radionuclide		Half-Life	kg/yr	Ci/yr
Thorium b/	228	1.910 yr	2.32×10^{-3}	1.91×10^{3}
	229	7340 yr	1.09×10^{-3}	2.32×10^{-1}
	230	8×10^{4} yr	2.64×10^{-1}	5.40
	231	25.5 hr	1.99×10^{-10}	1.06×10^{-1}
	232	1.41×10^{10} yr	6.75×10^{3}	7.38×10^{-1}
	234	24.1 day	2.46×10^{-5}	5.70×10^{2}
TOTAL			6.75×10^{3}	α 1.91×10^{3} β 5.70×10^{2}
Protactinium	233	27.0 day	3.65×10^{-1}	7.58×10^{6}
	234	6.75 hr	2.87×10^{-10}	5.70×10^{-1}
TOTAL			3.65×10^{-1}	β 7.58×10^{6}
Uranium	232	72 yr	1.39×10^{-1}	2.98×10^{3}
	233	1.62×10^{5} yr	1.89×10^{2}	1.79×10^{3}
	234	2.47×10^{5} yr	6.20×10^{1}	3.84×10^{2}
	235	7.1×10^{8} yr	4.91×10^{1}	1.05×10^{-1}
	236	2.39×10^{7} yr	1.03×10^{2}	6.53
	237	6.75 day	1.69×10^{-8}	1.38
	238	4.51×10^{9} yr	2.88×10^{1}	9.60×10^{-3}
TOTAL			4.32×10^{2}	α 5.16×10^{3} β 1.38
Neptunium	237	2.14×10^{6} yr	1.09×10^{1}	7.69
TOTAL			1.09×10^{1}	α 7.69
Plutonium d/	236	2.85yr	5.01×10^{-6}	2.66
	238	86 yr	5.62	9.82×10^{4}
	239	24,400 yr	1.18	7.24×10^{1}
	240	6,580 yr	5.54×10^{-1}	1.25×10^{2}
	241	13.2 yr	5.33×10^{-1}	5.99×10^{4}
	242	3.79×10^{5} yr	5.40x10-1	2.11
TOTAL			8.43	α 9.84×10^{4} β 5.99×10^{4}
Americium	241	458 yr	1.92×10^{-2}	6.22×10^{1}
	242 m	152 yr	3.01×10^{-4}	2.93
	243	7950 yr	1.55×10^{-1}	2.87×10^{1}
TOTAL			1.75×10^{-1}	α 9.09×10^{1} β 2.93
Curium	242	163 day	4.90×10^{-3}	1.62×10^{4}
	243	32 yr	1.30×10^{-4}	5.98
	244	17.6 yr	6.98×10^{-2}	5.81×10^{3}
	245	9300 yr	2.86×10^{-4}	4.49×10^{-2}
TOTAL			7.58×10^{-2}	α 2.20×10^{4}

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a/ 95 Mw day/kg HM, 38.7% thermal efficiency, 80% capacity factor 150-day cooling, equilibrium fuel cycle.

b/ Natural thorium is assumed to contain 100 ppm ^{230}Th. Discharge thorium is not recycled.

c/ Includes 59.0 kg/yr of second-cycle uranium from initial make-up ^{235}U, which is not to be recycled. Composition of discharged second-cycle uranium: 0.8% ^{234}U, 3.6% ^{235}U, 75.5% ^{236}U, 20.1% ^{238}U.

d/ Plutonium is not recycled.

Each of the three types of uranium in the fresh fuel is formed into microspheres from 570 to 580 microns in size, which are then mixed with 820-micron microspheres of thorium and embedded in a carbonaceous matrix to form a fuel "stick" (D1). The resulting fuel sticks are sealed into holes in blocks of high-purity nuclear-grade graphite, which acts as neutron moderator and structural support. Heat generated in the fuel sticks is conducted through the adjacent graphite into helium coolant, which flows through longitudinal holes penetrating each graphite fuel block.

Each fuel block contains only one of the three types of uranium-thorium fuel, so that the spatial arrangement throughout the reactor of blocks containing different types of fissile uranium provides a means of controlling the spatial distributions of neutron flux and power density.

The material properties of each of the three fuel types are given in Table 5.6. The inital and make-up fuel elements, containing the highly enriched (93.5%) make-up uranium, are formed by 200-micron microspheres of UC_2 and 500-micron microspheres of ThO_2. The uranium microspheres are each coated with an inner layer of low-density pyrolytic carbon to provide voids for fission products and to act as a buffer layer for fission-product recoil. Surrounding this is a layer of high-density pyrolytic carbon, a layer of silicon carbide, and then another layer of high-density pyrolytic carbon to reduce the diffusional escape of uranium and fission products from the fuel microspheres. The fuel elements of recycled ^{235}U and make-up thorium are formed from microspheres similar to those described above. In the fuel elements containing recycled ^{233}U and make-up thorium the uranium microspheres are formed from similar coatings of UC_2 particles initially 310 microns in diameter.

The steam generated by the hot helium coolant from the reactor is at higher temperature and pressure than the steam generated in water reactors, resulting in an over-all thermal efficiency of 38.7%. For a net electrical output of 1000 Mw the resulting thermal power is 2583.9 Mw.

The average thermal specific power in the reactor core is calculated to be 65.1 Mw per Mg of uranium and thorium in the fresh fuel. Each year one fourth of the reactor fuel, contained within 850 graphite fuel blocks (D1,L1) is discharged and replaced with unirradiated fuel, so that each fuel element remains within the reactor for four years. At an average load factor of 80% the resulting average thermal exposure is 95,000 Mw days per Mg of uranium and thorium charged (L1,T3).

The coatings surrounding the uranium and thorium fuel particles, as shown in Table 5.6, not only reduce the escape of fission products to the gas coolant during reactor operation, but they also aid the separation of fissile and fertile particles in fuel reprocessing. The reprocessing technique specified for HTGR fuel involves crushing and burning the graphite blocks in a fluidized combustor. The ash from the fluidized combustor consists of the

Table 5.6

HTGR Fuel Particle Descriptions (D1,L1,T3)

Property	^{235}U Make-up Elements[a]		^{233}U Recycle Elements		^{235}U Recycle Elements	
	Fissile Particle	Fertile Particle	Fissile Particle	Fertile Particle	Fissile Particle	Fertile Particle
Isotope	^{235}U	Th	^{233}U	Th	^{235}U	Th
Kernel Composition	UC_2	ThO_2	UC_2	ThO_2	UC_2	ThO_2
Kernel Diameter (μm)	200	500	310	500	200	500
Type Coating (b/,c/)	TRISO	BISO	TRISO	BISO	TRISO	BISO
Coating Thickness (μm)						
Buffer Carbon	100	85	50	85	100	85
Inner Dense Carbon	25		25		25	
Silicon Carbide (SiC)	25		25		25	
Outer Carbon	35	75	35	75	35	75
Total Particle Diameter (μm)	570	820	580	820	570	820

---

a/ For initial and make-up loadings

b/ A TRISO coating consists of a buffer layer surrounding the UC_2 kernel, followed by successive layers of dense pyrolytic carbon, silicon carbide, and dense pyrolytic carbon.

c/ A BISO coating consists of a buffer layer and a single layer of dense pyrolytic carbon.

original UC_2 particles still coated with silicon carbide and oxide particles of UO_2-ThO_2 from the incineration of the original ThO_2 particles coated with pyrolytic carbon. Although the sizes of these fertile and fissile particles are about the same after graphite combustion, the thoria particles are about three times heavier because of the larger diameter of their actinide kernel and because the SiC and inner carbon coatings of the fissile particles still remain. The fissile and fertile particles are separated into two fractions by elutriation with carbon dioxide. The thoria particles, now containing fission products and bred uranium, are to be processed by Thorex separation technology, and the fissile uranium particles containing recoverable uranium, fission products, and some neptunium and plutonium, are to be processed by Purex separation technology.

The purpose of making the size separation of the fissile and fertile particles from each block is to develop a means of controlling the build-up of neutron-absorbing ^{236}U. The fissile particles used to fabricate each graphite fuel block are one of three different types of uranium described in Table 5.6. Fuel blocks with different sources of fissile particles are to be processed separately through graphite combustion and particle classification, so that the three different groups of fissile particles can be collected and treated separately. The particles of uranium remaining from the first-cycle irradiation of make-up uranium are to be processed for uranium recycle. The particles of irradiated bred uranium are to be processed and the recovered uranium is to be combined with uranium recovered from thorium and recycled. The uranium particles remaining after the second irradiation cycle of initial make-up uranium contain a relatively high concentration of ^{236}U and are to be discarded to transuranic wastes.

Because elutriation does not produce a quantitative separation there will be some crossover of fertile and fissile particles, and the crossover will increase as a result of broken particles. The effects of crossover are to contaminate the recovered ^{233}U with ^{236}U neutron poison and to increase the loss of ^{233}U when fissile particles are retired. Upper-bound estimates (P4) indicate that as much as 10% of the fissile particles may cross over into the fertile stream although less actual crossover is expected. The calculated effects of crossover on the composition of the recycled uranium are shown in Table 5.7.

5.2 HTGR Fueled With Thorium and Denatured Uranium, No Reprocessing

Until facilities exist for reprocessing uranium-thorium HTGR fuel, any HTGR must operate on the non-reprocessing cycle, i.e., it must be fueled with low-enrichment uranium or with thorium blended with enriched ^{235}U or with plutonium recovered from LWR discharge fuel. The non-reprocessing fuel cycle of an HTGR fueled with thorium and enriched ^{235}U is shown in Figure 5.3. In this case, the isotopic content of ^{235}U in uranium is kept at 20% or below as a means of reducing the safeguards hazards associated with 93% ^{235}U in make-up uranium. These safeguards issues are

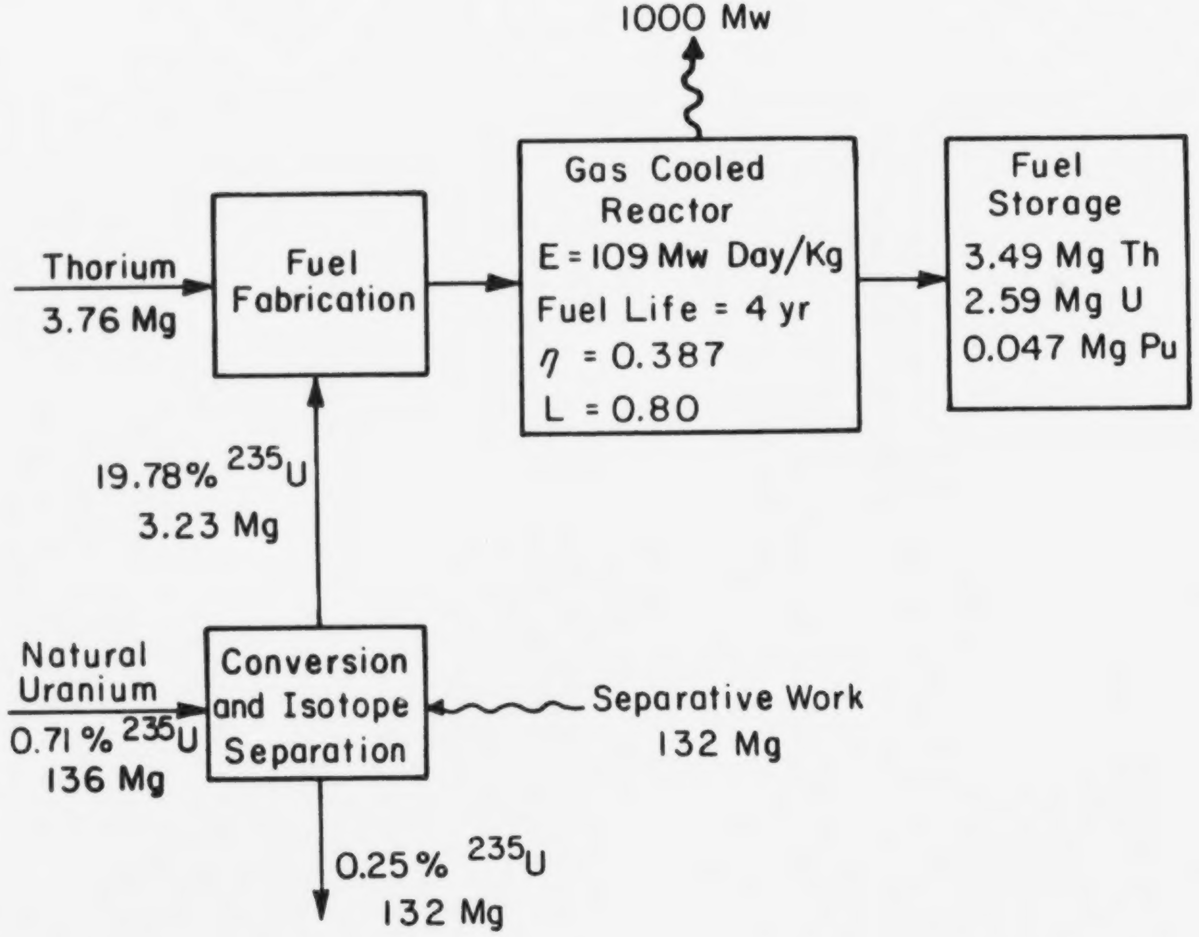
1000 Mw
Thorium
3.76 Mg
Fuel Fabrication
Gas Cooled Reactor
E = 109 Mw Day/Kg
Fuel Life = 4 yr
η = 0.387
L = 0.80
Fuel Storage
3.49 Mg Th
2.59 Mg U
0.047 Mg Pu
19.78% ^{235}U
3.23 Mg
Natural Uranium
0.71% ^{235}U
136 Mg
Conversion and Isotope Separation
Separative Work
132 Mg
0.25% ^{235}U
132 Mg

Fig. 5.3 Annual quantities for the denatured-U-Th-fueled HTGR, with no recycle (E=fuel capacity, η=overall thermal efficiency, L=capacity factor)

Table 5.7

Effect of Reprocessing Cross-Over on the Composition of Recycled Uranium For the HTGR Equilibrium Fuel Cycle

	No cross-over		With cross-over[a]	
	charge,%	discharge,%	charge,%	discharge,%
Recycled ^{235}U Particles	-----	------	-----	--------
^{232}U	0	0	0.002	0.001
^{233}U	0	0	4.17	0.39
^{234}U	1.22	0.81	1.99	1.74
^{235}U	29.51	3.96	28.15	4.08
^{236}U	52.24	75.28	49.53	74.17
^{238}U	17.03	19.95	16.16	19.61
Recycled Bred Uranium				
^{232}U	0.04	0.04	0.04	0.03
^{233}U	52.31	7.83	42.20	5.24
^{234}U	22.04	31.65	17.88	21.27
^{235}U	6.73	12.27	6.32	8.39
^{236}U	18.87	48.21	30.61	60.33
^{238}U	0	0	2.95	4.75

---

[a] Assumed reprocessing cross-over: 10% of the fissile particles into the fertile stream, 5% of the fertile particles into the fissile stream.

discussed in more detail in Chapter 7. Calculated material quantities for the near-equilibrium fuel cycle of the HTGR operating with denatured-uranium-thorium fuel, normalized to data of Haffner, et. al. (H3), are given in Tables 5.8 and 5.9.

5.3 Resource Utilization by Current and Modified HTGR Designs

The thirty-year lifetime ore requirements for the HTGR fueled with denatured-U-Th, with no recycle as in Fig. 5.3, and for the reference design of the HTGR, operating according to the overall flowsheet of Figure 5.1, are shown in Table 5.10. The non-reprocessing HTGR cycle requires 50% more uranium ore than does the U-Th reprocessing cycle of the HTGR reference design, but it requires 21% less uranium ore than does the uranium-fueled non-reprocessing cycle of the PWR (Fig. 3.1).

Also shown in Table 5.10 is an estimate for a similar HTGR which uses plutonium recovered from LWR discharge fuel as make-up fissile material (P2). Both uranium and plutonium in this version of the HTGR are assumed to be recycled. The ore required for this case includes ore for a uranium-fueled LWR to supply the make-up plutonium, with the total reactor system scaled to an electrical energy generation of 30x0.8 Gw yr. Much of the ore required for these recycle cases is that necessary to supply the fissile material for the initial loading and for extra fissile make-up during the many irradiation cycles before the near-equilibrium fuel cycles are reached. However, a second-generation HTGR could start-up with the fuel cycle inventory left from a retired first-generation HTGR. The lower lifetime ore requirements of such second-generation HTGR's are shown in Table 5.10. The HTGR can also be operated on low-enrichment uranium, with or without U-Pu recycle.

Comparing the data in Tables 3.1, 4.1 and 5.10 for thorium fuel cycles with ^{235}U make-up, it is apparent that the current HTGR reference design is intermediate in ore requirement between the less efficient thorium version of the LWR and the more efficient thorium versions of the CANDU reactor.

TABLE 5.8

Actinides in the Fuel Charged to the Denatured HTGR

(1000 MWe, Stowaway cycle[a])

Radionuclide	kg/yr	Ci/yr[b]	weight %
Thorium 232	3.72×10^{3}	4.07×10^{-1}	100.00
total	3.72×10^{3}	$\alpha = 4.07 \times 10^{-1}$	100.00
Uranium 235	6.33×10^{2}	1.36	19.78
238	2.57×10^{3}	8.55×10^{-1}	80.22
total	3.20×10^{3}	$\alpha = 2.22$	100.00

---

[a] 109 Mw day/kg U+Th, 38.7% thermal efficiency, 80% capacity factor, near-equilibrium fuel cycle

[b] "α" denotes radioactive decay by alpha emission

TABLE 5.9

Actinides In The Fuel Discharged From The Denatured HTGR
(1000 MWe, Stowaway cycle[a])

Radionuclide		kg/yr	Ci/yr[e]	weight %
Thorium[b]	232	3.49×10^{3}	3.81×10^{-1}	100.00
total		3.49×10^{3}	$\alpha = 3.81 \times 10^{-1}$	100.00
Uranium[c]	233	8.50×10^{1}	8.06×10^{2}	3.28
	234	1.60×10^{1}	9.90×10^{1}	0.62
	235	5.50×10^{1}	1.18×10^{-1}	2.12
	236	8.70×10^{1}	5.52	3.36
	238	2.35×10^{3}	7.82×10^{-1}	90.62
total		2.59×10^{3}	$\alpha = 9.11 \times 10^{2}$	100.00
Plutonium[d]	239	1.70×10^{1}	1.04×10^{3}	36.17
	240	1.00×10^{1}	2.27×10^{3}	21.28
	241	8.00	9.00×10^{5}	17.02
	242	1.20×10^{1}	4.68×10^{1}	25.53
total		4.70×10^{1}	$\alpha = 3.36 \times 10^{3}$ $\beta = 9.00 \times 10^{5}$	100.00

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[a] 109 Mw day/kg U+Th, 38.7% thermal efficiency, 80% capacity factor, near-equilibrium fuel cycle. Np, Am, Cm are not included.

[b] Other thorium isotopes are not included.

[c] ^{232}U and ^{237}U are not included.

[d] ^{238}Pu is not included.

[e] Activities from alpha decay and beta decay are indicated by "α" and "β", respectively.

TABLE 5.10

30-Year Lifetime Ore Requirements for High-Temperature Gas-Cooled Reactors
(Current HTGR Reference Design, 1000 Mw Electrical Power, 80% Capacity Factor)

Fuel Cycle	Conversion Ratio	Natural Uranium, Metric Tons (Mg)		Natural Uranium relative to U-fueled PWR w/o recycle[a]	Thorium, metric tons
		0.20% depleted U	0.25% depleted U		
(a) Denatured-U-Th fuel, no recycle	0.50	3870	4270	0.79	151
(b) ^{235}U-Th fuel, U recycle	0.66	2290	2840	0.53	247
(c) Pu-Th fuel, U-Pu recycle[d]	0.64	3850	4130	0.77	79.9
(d) 2nd gen. ^{235}U-Th, U recycle	0.66	1920	2130	0.40	221
(e) 2nd gen. Pu-Th, U-Pu recycle[e]	0.64	1974	3000	0.56	70.3

---

[a] 30.4 Mw-day/kg HM, 34.2% thermal efficiency, 80% capacity factor, near-equilibrium cycle.

[b] ^{234}U is not included.

[c] 150 days cooling of discharged fuel before reprocessing. 1.5% loss in reprocessing, 1.5% loss in fabrication. Natural uranium is added to the recycled plutonium to dilute the recycle fuel to proper enrichment.

[d] Includes U-fueled PWR to supply make-up Pu. Total system operates for 30 yr at 1000 Mw.

[e] Starts with equilibrium reactor and fuel-cycle fissile inventory.

The current reference HTGR, which is the basis of the flowsheet of Figure 5.1 and the data in Table 5.10, is a design optimized for current or near-term fuel-cycle cost parameters, including uranium ore prices. However, a feature of the HTGR fuel concept is the flexibility for making relatively large changes in the fuel loading without altering the fuel thermal performance or the overall mechanical design, or with only modest changes in these design parameters. For a future era of higher uranium ore prices and greater incentive to improve ore utilization, the conversion ratio of the HTGR can be increased by the following modifications (B5):

1. Increase the thorium loading in the core, which increases neutron absorption in thorium relative to non-productive absorption and leakage.

2. Decrease the core power density. For the same fissile concentration, this decreases the neutron flux and reduces the flux-dependent neutron absorption in ^{135}Xe and ^{233}Pa. The greater core volume, for the same thermal power, provides volume for further increases in the thorium loading.

3. Decrease the time interval between refueling, thereby decreasing the loss of neutrons to control absorbers.

4. Reduce the thickness of the coatings on the fuel particles, allowing greater theorium loading in the graphite-prism fuel holes and thereby allowing greater thorium loading per prism.

5. Distribute the fuel particles uniformly throughout the graphite prism.

Estimated improvements in the conversion ratio and ore requirements made possible by such approaches are shown in Table 5.11.

The first two modifications, which increase the conversion ratio to 0.76, can be achieved with the current fuel element design. This reduces the life-time ore requirement to a level about 58% greater than that of the CANDU reactor operating on the same fuel cycle.

The most significant parameter in increasing the HTGR conversion ratio is the increased thorium loading. This requires corresponding increases in the initial and start-up loadings of fissile ^{235}U, resulting in a greater investment in fuel early in the reactor life. The higher initial investment contributes to a higher levelized fuel cycle cost, but if the price of uranium increases more rapidly than does the effective discount factor during the plant life, the lower annual ore requirements for fuel reloads throughout the plant life could possibly offset this higher initial fuel investment.

One feature of the HTGR which benefits its fuel cycle cost and its resource utilization is the very high fuel exposure of 94 Mw day per kilogram of heavy metal. This means that for fuel reprocessing and refabrication to make the same contribution to the cost of electrical

TABLE 5.11

Conversion Ratio Improvements Possible for the HTGR Fueled with ^{235}U, Th, and Recycled Uranium

Modification[a]	Conversion ratio[a]	Lifetime ore requirement[b]	
		relative to reference HTGR	relative to U-fueled PWR w/o recycle[c]
Reference HTGR, ^{235}U-Th fuel, U recycle	0.66	1.0	0.53
Increases thorium loading by 25%	0.71	0.89	0.47
Change from yearly fueling to semiannual fueling	0.76	0.80	0.42
Reduce core average power density from 8.4 to 6.0 watts/cm^3	0.82	0.68	0.36
Use modified fuel elements	0.90	0.57	0.30
and/or improved fuel particles	0.95	-----	-----

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[a] From Brogli, et al., (B4).

[b] Includes initial loading and reloads over 30-yr lifetime, calculated from data of Brogli, et al., (B4).

[c] 30.4 Mw-day/kg HM, 34.2% thermal efficiency, 80% capacity factor, near-equilibrium cycle.

energy, the unit costs of these operations, expressed in cost per unit amount of heavy metal processed, can be correspondingly greater for these operations in the HTGR fuel cycle. However, whether these HTGR operations can be carried out within the greater allowable unit costs is uncertain at this time. The HTGR reprocessing operations are yet to be carried out on a pilot-plant scale, so the technological foundation for estimating the cost of commerical-scale operations is now quite limited.

The burnup advantage of HTGR fuel over LWR and CANDU fuels decreases, but does not disappear, as modifications are made to improve the HTGR conversion ratio. The improvements in conversion ratio and ore requirements listed in Table 5.10 were calculated on the assumption that the fuel would be irradiated for a constant time interval of four years, as in the present HTGR reference design. Therefore, as the thorium loading and fissile loading are increased to improve the conversion ratio, the burnup correspondingly decreases.

The HTGR reactor design is well founded and is readily adaptable to the modifications described herein. However, the technical complexities and lack of engineering-scale experience in the HTGR fuel cycle suggest caution in economic comparisons with other fuel cycles. Thorough and periodic engineering evaluation of the economics of the HTGR fuel cycle and of alternative thorium-based fuel cycles is important. Similar reactors are under development in Germany, where designs of the prismatic type as well as advanced pebble bed designs are being considered.

6. Fuel Cycles for Fast-Breeder Reactors*

6.1 The Reference PuO_2-UO_2 LMFBR

Consideration of fast-breeder fuel cycles is relevant to the issues of thorium fueling because:

(a) The possible resource need for a thorium fuel cycle alternative in near-term reactors to reduce the consumption of uranium ore disappears if and when the uranium-plutonium breeders are introduced. However, if the uranium-plutonium breeder is deferred or delayed, an alternative thorium fuel cycle in LWR, HTGR, or CANDU reactors may become an important means of conserving uranium resources.

(b) Fast breeders fueled with metallic thorium and recycled uranium can achieve higher breeding ratios and lower sodium-void coefficients of reactivity than PuO_2-UO_2 fast breeders.

(c) Fast breeders with U-Pu cores and blanketed in part with thorium can consume the plutonium produced by dispersed national reactors fueled with denatured uranium and thorium and can supply the make-up ^{233}U to fuel these safeguarded national reactors. This is considered in more detail in Chapter 7.

The fast-breeder programs in this country and elsewhere are focused on the development of the sodium-cooled breeder reactor fueled with PuO_2 and UO_2. In a given type of fuel material, such as oxide fuel, higher breeding ratios and shorter doubling times are possible with the uranium-plutonium fuel to start up the first generation breeders. Moreover, the Purex reprocessing technology is available for the uranium-plutonium cycle. Figure 6.1 shows a flowsheet for a possible early LMFBR operating on an equilibrium fuel cycle (G1) fueled with natural or depleted uranium. The excess plutonium production from this breeder can be used to start up subsequent breeders, provided that the doubling time for increasing breeder capacity is no shorter than the doubling time for excess fissile production by the breeder.

A large amount of depleted uranium from isotope separation will have been stockpiled by the time when present low-cost uranium resources are consumed by water reactors. Assuming that breeders replace the water reactors then being retired in the next century, and assuming that the total fission power continues at a constant level, the stockpiled depleted uranium is an already-mined resource sufficient to fuel these breeders for thousands of years. The fast breeder is the most efficient of all fission systems in terms of long-term ore utilization.

The only ore requirement attributable to the breeder is that associated with the production of plutonium for start-up loadings of the first-generation breeders. This plutonium must be obtained

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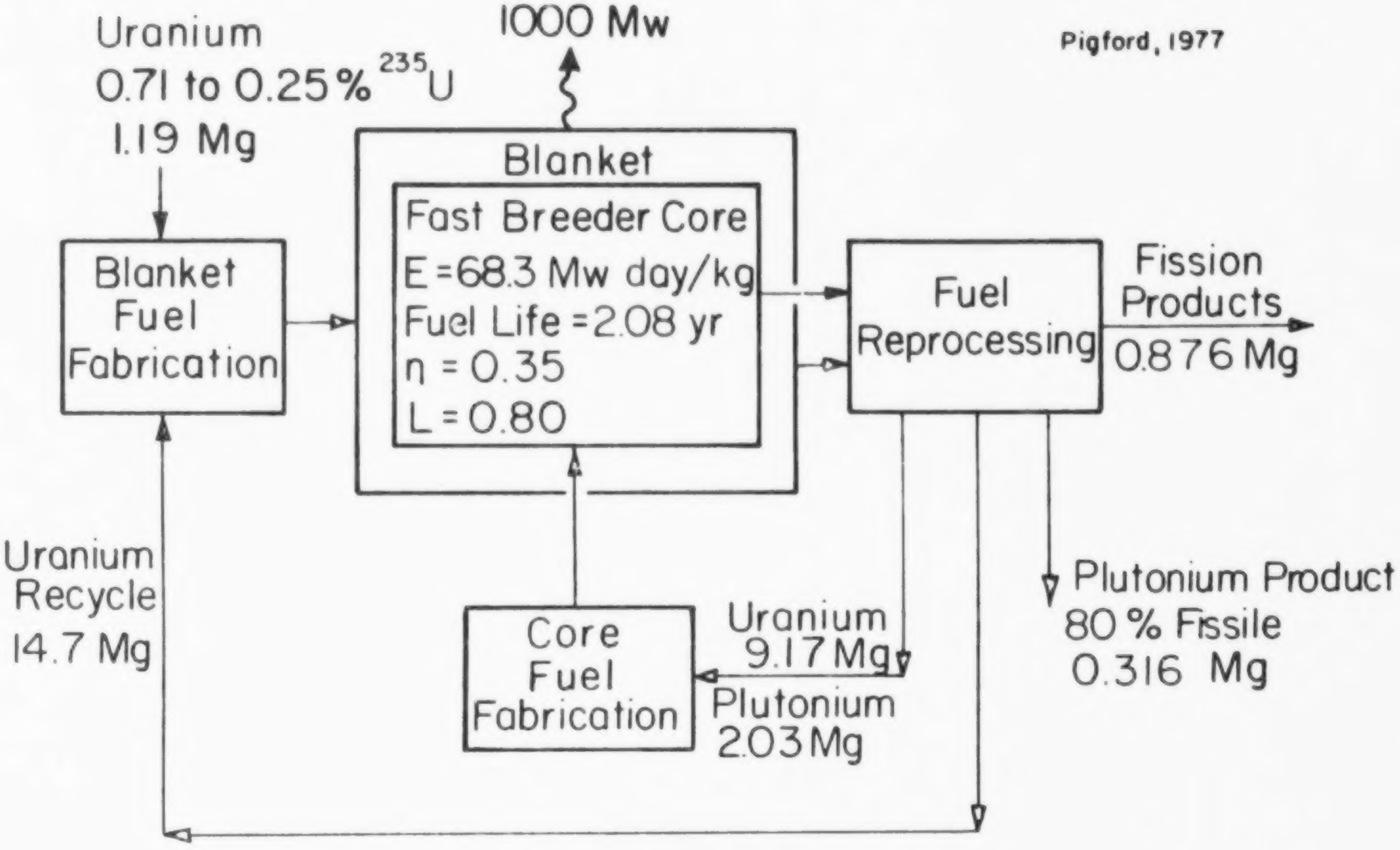
*Much of the text of this section was adapted by the first author for incorporation in the APS report on nuclear fuel cycles and waste management (H1).


Uranium
0.71 to 0.25% ^{235}U
1.19 Mg
1000 Mw
Blanket Fuel Fabrication
Blanket
Fast Breeder Core
E = 68.3 Mw day/kg
Fuel Life = 2.08 yr
η = 0.35
L = 0.80
Fuel Reprocessing
Fission Products
0.876 Mg
Uranium Recycle
14.7 Mg
Core Fuel Fabrication
Uranium
9.17 Mg
Plutonium
2.03 Mg
Plutonium Product
80% Fissile
0.316 Mg

Pigford, 1977

Fig. 6.1 Annual quantities for LMFBR fueled with natural or depleted uranium (equilibrium fuel cycle, E=fuel exposure, η=thermal energy, L=capacity factor)

from light-water reactors, and these reactors will then require more ore because they are thereby deprived of the benefits of plutonium recycle. When operating without Pu recycle, the 1-Gw LWR produces 171.4 kg/yr of fissile Pu. LWR's must operate for 43.8 Gw-yr without Pu recycle in order to produce the 7500 kg of fissile Pu required to start-up a 1-Gw LMFBR. The uranium ore attributable to Pu production is the difference between the ore required for operating LWR's with U recycle only and that required with U-Pu recycle. Using the data of cases (b) and (c) of Table 3.3 and scaling to 43.8 Gw-yr, we estimate 1180 Mg of natural uranium, for 0.25% depleted uranium tails attributable to Pu start-up as shown in Table 6.1.

If the doubling time for subsequent growth in breeder capacity is no greater than the doubling time for the breeder to produce excess plutonium, no ore is required for subsequent breeder generations. For each gigawatt of first-generation breeder capacity installed in the 1990's, 43.8 Gw-yr of light water reactors must be operated without plutonium recycle during the 1980's and early 1990's to furnish the start-up plutonium. Therefore, breeder introduction in the 1990's would require the existence of industrial-scale LWR reprocessing several years before that time. The present schedule is uncertain.

The data in Table 6.1 indicate that over a 30-year operating life, three uranium-fueled light water reactors could produce enough plutonium to start up two fast breeders, if no plutonium were to be recycled in water reactors. Alternatively, nine water reactors operating during their last ten years of life without plutonium recycle will generate enough plutonium to eventually start up two breeders. The 1974 ERDA projections of U.S. nuclear power growth indicated a growth to 124 GW of fast breeder capacity by the end of the century, along with 644 GW of light water reactors. Calculations (P2) of the amount of start-up plutonium required for such a large scale of breeder introduction showed that plutonium recycle in water reactors would have to be discontinued in the early 1990's to insure sufficient plutonium for breeder start-up. However, events since 1974 suggest that such a rapid introduction of breeders is not likely, and delays in LWR fuel reprocessing and in the construction of additional LWR fuel reprocessing facilities seem more likely to result in an over supply in the 1990's of plutonium which can be extracted from water reactor fuel.

From the above it is apparent that there are several situations, any one of which could warrant operating water reactors entirely with uranium fueling so that all of the plutonium produced would be available for breeder start up. Examples are:

TABLE 6.1. Fissile, Ore, and Enrichment Requirements to Start a First-Generation Fast Breeder Reactor with Water-Reactor Plutonium (1000 Mw electrical power, 80% capacity factor).

Fissile Pu required for fast breeder start-up[a]	7500 kg
Operation of U-Fueled water reactor to generate Pu start-up inventory	43.8 Gw Yr
U in ore attributable to production of startup Pu	
0.20% depleted U	1060 Mg natural U
0.25% depleted U	1180 Mg natural U
Additional separative work due to loss of Pu-recycle in water reactors:	
0.20% depleted uranium	1200 Mg
0.25% depleted uranium	1020 Mg

Example: To start up 1 GW of FBR requires that 4.38 Gw of LWR be operated for 10 yr. without Pu recycle. Total natural U used = 6540 Mg, assuming 0.25% depleted U. Total natural uranium attributable to breeder start-up = 1180 Mg.

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[a] Based upon 3000 kg fissile Pu for the initial core plus 4500 kg for replacement loadings before Pu in discharge fuel is recycled (G1).

(a) a very limited supply of uranium ore

(b) a sufficiently large ratio of first-generation breeders to previously installed water reactors

(c) a desire to move as rapidly as possible into a breeder power system.

6.2 Fast Breeder Start-up with ^{235}U

Plutonium is the best of all the fissile isotopes in achieving high breeding ratios and low doubling times in fast breeders (B12, G1, P9, Y1, Y2). Although enriched (20 to 22%) ^{235}U from isotope separation could be used for breeder start-up, the relative penalties associated with ^{235}U results in larger fissile inventory and a lower breeding ratio than with plutonium fueling.

Calculated amounts of natural uranium and separative work for ^{235}U start-up are presented in Table 6.2. It is shown that for a commercial fast breeder optimized for an equilibrium plutonium-uranium fuel cycle, the amount of fissile uranium required for start-up is from 1.5 to 2.4 times as large as the amount of fissile plutonium that would be required, depending upon the method of reprocessing the core fuel. ^{235}U start-up would consume greater quantities of uranium ore than that attributable to Pu start-up from LWR's, and also would require greater quantities of electrical energy for isotope separation. The corresponding total cost of the fissile material for start-up would be greater by a factor of 2.3 to 3.7 for enriched uranium than for plutonium (H1, P9).

The breeding ratio is significantly lower during start-up cycles with ^{235}U, and this effect persists for many subsequent reloads until most of the ^{235}U has been recycled and consumed. The new deficit in breeding-gain production of fissile plutonium due to ^{235}U start-up of a 1000 Mw LMFBR is about 1700 kg. This considerably increases the breeding-gain doubling time and will delay the start-up of second-generation breeders, assuming these are to be fueled initially with Pu from first-generation breeders.

6.3 Summary of Resource Requirements for the Reference LMFBR

The total lifetime ore requirements for the reference LMFBR, including the ore for start-up and the ore for life-time refueling, are shown in Table 6.3. For the first-generation breeders, which require start-up fissile material from an external non-breeder source, the lifetime ore requirement is still less than any of the first-generation light-water-reactor cases listed in Table 3.3 and is less than any of the first-generation HTGR cases listed in Table 5.5, when all cases are calculated with the same concentration of ^{235}U in depleted uranium from isotope separation. However, any one of the first-generation CANDU cases with recycle (Table 4.1) requires less lifetime ore than does the LMFBR with ^{235}U start-up but requires more lifetime ore than does the first-generation LMFBR with plutonium start-up. The second-generation LMFBR, which received its start-up fissile inventory from first-generation breeders, requires no ore if it is fueled with depleted uranium which has been previously stockpiled from isotope separation. If fueled with natural uranium, the

TABLE 6.2. Fissile, Ore, and Enrichment Requirements to Start A First-Generation Fast Breeder Reactor on Enriched Uranium (1000 Mw electrical power, 80% capacity factor).

A. Uranium in discharge core fuel is reproduced separately from uranium in axial and radial blankets	
Fissile ^{235}U required for fast breeder start-up[a], Mg	11.25
Natural uranium in ore required for 20% ^{235}U, Mg	
0.20% depleted uranium	2160
0.25% depleted uranium	2400
Separative work required for 20% ^{235}U, Mg	
0.20% depleted uranium	2560
0.25% depleted uranium	2330
B. Fuel elements containing core and axial blanket are chopped and processed without core-blanket separation, so enriched uranium is not recycled	
Fissile ^{235}U required for fast breeder start-up[a], Mg	18.00
Natural uranium in ore required for 20% ^{235}U, Mg	
0.20% depleted uranium	2480
0.25% depleted uranium	3840
Separative work required for 20% ^{235}U, Mg	
0.20% depleted uranium	4100
0.25% depleted uranium	3730

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[a] Based upon 4.5 Mg ^{235}U for the initial core plus sufficient replacement loadings until reactor is self-sustaining on recycle fissile material. Although lower ^{235}U loadings are possible for a breeder core optimized for ^{235}U fueling, the purpose here is to start-up a core optimized for steady-state fueling on bred plutonium (G1, P9).

TABLE 6.3. 30-Year Lifetime Ore and Enrichment Requirements for Fast-Breeder Reactors[a] (1000 Mw electrical power, 80% capacity factor)

	First-generation breeder		Second-generation breeder
Source of fissile material for start-up	Pu from U-fueled LWR	20% enriched U [b]	Pu from first-generation breeder
Amount of start-up fissile material, Mg	7.5	11.25	7.5
Natural uranium in ore to produce start-up fissile inventory, Mg	1180	2400	0
Separative work attributable to breeder start-up, Mg	1020	2330	0
Natural uranium in ore for inventory and lifetime refueling			
if fueled with natural uranium, Mg	69.3 [c]	69.3[c]	35.7
if fueled with depleted uranium from stockpile, Mg	0	0	0
Total natural uranium in ore for start-up and for lifetime refueling			
if fueled with natural uranium,			
Mg	1250	2470	35.7
relative to lifetime requirement for U-fueled PWR	0.226	0.454	0.007
if fueled with depleted uranium from stockpile,			
Mg	1180	2400	0
relative to lifetime requirement for U-fueled PWR	0.220	0.447	0

[a] Calculated for 0.25% ^{235}U in depleted uranium from isotope separation.

[b] Increase material quantities by 60% if core and axial blanket are chopped and processed without core-blanket separation, so enriched uranium is not recycled (P9).

[c] Includes start-up inventory of uranium in reactor and fuel cycle. Assume two-year hold-up in external fuel cycle.

lifetime ore requirement of the second-generation LMFBR is less than 1% of that of the best second-generation, i.e., equilibrium inventory, cases of the PWR of Chapter 3; it is 1.7% of the lifetime ore requirement of a second-generation reference-design HTGR of Chapter 5; it is about 4% of the lifetime ore requirement for the best second-generation CANDU case of Chapter 4.

Any other breeder, such as thermal breeders that may result from modification of the thermal reactors discussed herein, can ultimately operate on an equilibrium fuel cycle that requires no greater ore for refueling than does the second-generation LMFBR. However, the time to reach equilibrium for these thermal breeders is greater than for the LMFBR, and there will be a greater ore consumption by the thermal breeders before breeding equilibrium is reached.

6.4 Thorium Fuel Cycles For Fast Breeder Reactors

There is some interest in breeders fueled with thorium and recycled uranium. For example, if the breeder program were to be significantly delayed and if thorium fueling of thermal reactors were to be introduced, as discussed earlier, to conserve uranium resources, these thermal reactors would eventually become sources of ^{233}U instead of plutonium for breeder start-up. Although ^{233}U is far better than ^{235}U for this purpose and results in reasonable breeding ratio, it is still inferior to plutonium.

When ThO_2 is substituted for UO_2 in the core fuel, case (b), the breeding ratio decreases. This is a result of the lower fast fission cross section of ^{232}Th and also from the partial replacement of ^{239}Pu by ^{233}U as the latter builds up and fissions during the irradiation cycle. Substituting ThO_2 for UO_2 in the blanket only slightly decreases the breeding ratio because of the relatively few fissions in the blanket. A core fueled with $^{233}UO_2$ - ThO_2 results in an even lower breeding ratio.

Since the irradiation behavior of UO_2 - ThO_2 fuel appears to be similar to that of PuO_2 - UO_2 fuel, it is likely that LMFBR's designed and introduced with PuO_2 - UO_2 fueling could be converted later to UO_2 - ThO_2 fueling. A longer doubling time would result, but the extent to which this would be a problem would depend upon the desired rate of breeder introduction.

Uranium-thorium fueling in breeder cores may have some safety advantage because of the smaller increase in reactivity from sodium voiding than with plutonium-uranium fueling. However, there are other means of reducing the reactivity effects of sodium voiding, if this proves to be necessary in the LMFBR development program. Introducing thorium fueling in breeders would introduce many of the problems that would be encountered with thorium fueling in thermal reactors. The build-up of ^{232}U in the irradiated fuel and the high-energy gammas of the ^{232}U-decay daughters would require more shielding and remote handling in fabricating recycled fuel, and it complicates fuel reprocessing. The ^{232}U build-up in a thorium-fueled fast reactor is likely to be considerably greater than in thorium-fueled thermal reactors. Also, the reprocessing would have to be based upon Thorex technology, which is not as well developed as Purex reprocessing and is expected to be more difficult and expensive. The control of shut-down reactivity is more difficult with ^{233}U fuel because of the relatively long (27.0 day) half life of ^{233}Pa, the ^{233}U precursor. The long half life results in increased precursor concentrations during operation. Significant reactivity is added by ^{233}Pa decay after reactor shutdown, and more control absorbers are required with uranium-thorium fueling. Also, the delayed neutron fraction for ^{233}U is lower than that for ^{239}Pu, so lower worth for individual control absorbers and slower withdrawal rates to avoid prompt criticality may be required. These operational problems can all be accommodated through proper design, but they can affect the economics of uranium-thorium fueling.

More advanced sodium-cooled breeders designed for higher breeding ratios and higher specific powers may be based upon fuel materials in the form of carbides, nitrides or metals. As shown in Table 6.4, these advanced fuel materials offer better theoretical thermal and neutronic performance, but less is known about their irradiation behavior than is known about oxide fuels. Also ^{14}C formation in nitride fuels may result in greater expense in environmental controls and in waste management. Although uranium-metal fuels have been considered unacceptable for the high burnups required for breeder cores, experience of the EBR-II experimental breeder now indicates that alloyed uranium metal may be suitable. Fuels of thorium-base alloy may be an even more attractive possibility. The isotropic face-centered-cubic structure of thorium metal is more stable than uranium to irradiation damage and swelling (S2). Thorium undergoes its solid-phase transformation at 1365°C, which is much higher than the 660°C transformation temperature of uranium metal. Also thorium melts at 1725°C, as compared with 1132°C for uranium. However, because of the limited solubility of uranium and plutonium in thorium, the irradiation behavior of U-Th and U-Pu-Th alloys for core fuel may not be as good as that expected for thorium metal. The irradiation behavior of such alloys at operating temperatures and design burnups is not sufficiently known.

The higher thermal conductivity of thorium-based alloys could result in higher specific power than with oxide fuel. Also, the higher atomic density of the metal should result in a breeding ratio higher than that attainable with oxides, as shown by cases (d) and (e) in Table 6.4. The higher specific power and breeding ratio both result in a lower doubling time for the thorium-alloy fuel. Also, with metal fuel the reactivity effects from sodium voiding are further reduced below those predicted for the oxides. These possible advantages from thorium-alloy fuel in breeders, as compared to thorium oxide fuel, must be weighed against the greater uncertainties in irradiation behavior and possibly more expensive fuel fabrication. Also, thorium alloy

TABLE 6.4. Comparison of Pu-U and U-Th Fueling in LMFBR's [a]
(1000 Mwe, 0.8 load factor).

			Excess Fissile Production kg/Gw yr				
Core Fuel Material	Blanket Material	Breeding Ratio	^{233}U	^{235}U	^{239}Pu	^{241}Pu	net
Mixed-Oxide Fuels							
a) $^{239}PuO_2$-UO_2	UO_2	1.23	---	31.3	165.1	7.6	141.4
b) $^{239}PuO_2$-ThO_2	UO_2-ThO_2 [b]	1.15	43.3	---	-34.6	3.8	96.5
Metallic Fuels							
c) ^{239}Pu^{233}U-Th	U-Th [c]	1.31	335.8	---	-104.9	1.7	232.6
d) ^{233}U-Th [c]	U-Th	1.21	-44.1	----	210.1		166.0

---

[a] Calculated for equilibrium fuel cycle (S2).

[b] Depleted UO_2 radial blanket, ThO_2 axial blanket.

[c] Metal core, depleted U metal radial blanket, Th metal axial blanket.

fuels will be subject to the same problems of thorium technology described above. Therefore, the present state of knowledge on thorium fueling in fast breeders does not suggest diversion from the PuO_2 - UO_2 fuel now under development. Advanced carbide and thorium-alloy fuels do offer promise for longer-range improvements in advanced breeder designs.

Thorium-alloy fuels for breeder cores are not adaptable to the concept of a breeder fueled with denatured uranium for safeguards fuel cycles. If the recycled ^{233}U were diluted by natural uranium to a fissile content of 12 to 20% (see Chapter 7), as is proposed for denatured uranium fuel cycles, core reactivity limitations do not allow further dilution with thorium. Also, even for an all-uranium ^{233}U- ^{238}U denatured core, it will be difficult to reach criticality if the ^{233}U content must be kept as low as 12%, as suggested in Chapter 7.

Helium-cooled fast breeders have also been studied and are still receiving research and development support. Higher breeding ratios are theoretically possible. However, less is known about the structural stability of the fuel and the irradiation behavior of fuel cladding. Also, approaches to emergency cooling which differ from those designed for the LMFBR are necessary.

7. Technical Safeguards Features of Thorium Fuel Cycles and Denatured Fuel Cycles

7.1 Safeguards in Normal Thorium Fueling

"Normal" thorium fueling consists of thorium mixed with highly enriched fissile make-up and recycled uranium, as has been illustrated in Sections 3, 4 and 5. For the equilibrium fuel cycles of a uranium-thorium fueled light water reactor, the recycled uranium typically contains about 55% ^{233}U and 10% ^{235}U, which is a fissile content sufficient for nuclear explosives. However, the recycled uranium will contain appreciable concentrations of ^{232}U. As illustrated in Figure 2.3, the gamma activity and external gamma dose rate due to ^{232}U daughters grows rapidly after fuel reprocessing. After 100 days a metallic uranium part as small as one kilogram and containing 100 ppm ^{232}U will produce a gamma dose rate as large as 0.1 rem/hr at one meter. Recycled uranium in a uranium-thorium cycle may contain several hundred to several thousand ppm of ^{232}U, depending upon the ^{230}Th content of the make-up thorium and upon the fuel lattice, so the surface dose rate will be considerably greater than shown in Figure 2.3. Therefore, recycled uranium from thorium irradiation will require more shielding than reactor-grade plutonium. This could affect the practicality of using ^{233}U-rich recycled uranium for explosives.

The fissile make-up for normal thorium fueling, as illustrated in Sections 3, 4, 5, consists either of highly enriched (93%) ^{235}U or plutonium recovered from discharge fuel from uranium-fueled water reactors. Highly enriched ^{235}U is the least radioactive of all the separated fissile materials. It can be handled with relatively little hazard from its radioactivity. Although its fast-assembly critical mass is greater than that of plutonium, ^{235}U has a relatively low neutron background from spontaneous fission and from (α,n) reactions. It can be assembled into simple gun-type devices. Uranium metal is less reactive chemically than plutonium metal. Therefore, the use of highly enriched ^{235}U introduces what may be the most significant of all the safeguards concerns in the various nuclear fuel cycles.

If thorium fuels are reprocessed soon after discharge from the reactor, appreciable quantities of undecayed ^{233}Pa may be present. A relatively simple chemical separation could yield pure ^{233}Pa. Its subsequent decay to ^{233}U would yield a pure fissile material for explosives.

7.2 Low-Enrichment Denatured-Uranium Fuel Cycles

Various fuel cycles have been suggested as means of restricting the possibilities of diverting fissionable material from nuclear power fuel cycles. The non-reprocessing fuel cycle for a uranium-fueled light water reactor, as illustrated in Figure 3.1, could be adapted to an international safeguard fuel cycle. The low-enrichment uranium fuel, containing about 3% ^{235}U, is "denatured" in the sense that additional isotopic enrichment would be required for it to be used as material for a nuclear explosive. The discharge fuel, which contains significant quantities of fissionable plutonium, could be stored under international inspection or control (i.e., an international stowaway cycle). This cycle will ultimately entail higher costs, since it is the greatest consumer of natural uranium and requires a relatively large supply of slightly enriched uranium. The alternative of reprocessing the discharge fuel and storing the recovered plutonium under international inspection or control may impose additional safeguards and financial burdens. The stored plutonium must be protected, and the cost of storing separated plutonium is high compared with the cost of storing discharge fuel (H1,P3). Nevertheless, the stowaway cycle is technically the simplest of the alternatives discussed herein and can be consistent with their later implementation. If such international safeguards fuel cycles are to be utilized, the stowaway version represents a possible first step that could be implemented with existing technology.

Another alternative is to fuel all such national reactors, to be under individual safeguard control, with slightly enriched uranium and to ship the discharge fuel to a centralized fuel reprocessing center under international control. The recovered plutonium would be consumed on-site in plutonium-burner reactors. The electrical distribution system receiving the energy generated by plutonium-burner reactors would require relatively little uranium ore. The uranium ore thus saved could then be used as feed to a centralized uranium-enrichment plant to supply the slightly enriched uranium fuel for the externally located uranium-fueled reactors. The total uranium ore consumption for the entire generating system would be the same as if all reactors were nationalized and operating with self-generated uranium-plutonium recycle. However, financial and uranium exchanges between participating countries are required. An overall flowsheet of this safeguards fuel cycle at equilibrium is shown in Figure 6.1. Since the fuel discharged from the uranium-fueled reactors would still contain plutonium, the storage and shipment of the discharge fuel would have to be under safeguard control. Again, this cycle represents a step based on an existing technology and could be implemented in the near future.

In calculating the actinide quantities for the national-international fuel cycles shown herein, it has been assumed that 1% of the actinides are lost to reprocessing wastes and 1% to fabrication wastes. It is obviously necessary that the fissile content in these wastes be identified and safeguarded.

7.3 Denatured-Uranium-Thorium Cycles with Pressurized-Water Reactors

An alternative to the uranium cycles is the thorium-uranium cycle, in which ^{233}U is formed by neutron absorption in ^{232}Th. The fissile uranium in the fuel is to be denatured by dilution with natural or depleted uranium. The isotopic concentrations at which the fast-assembly critical masses for ^{235}U-^{238}U and ^{233}U-^{238}U mixtures become very large, and presumably unsuitable for explosives, are (H1, P5):

$$\frac{^{235}U}{^{235}U + {}^{238}U} = 0.20$$

$$\frac{^{233}U}{^{233}U + {}^{238}U} = 0.12$$

For fuels containing masses M_{233}, M_{235}, and M_{238} of ^{233}U, ^{235}U, and ^{238}U, respectively, the required dilution by ^{238}U is assumed to be obtained by the linear combination:

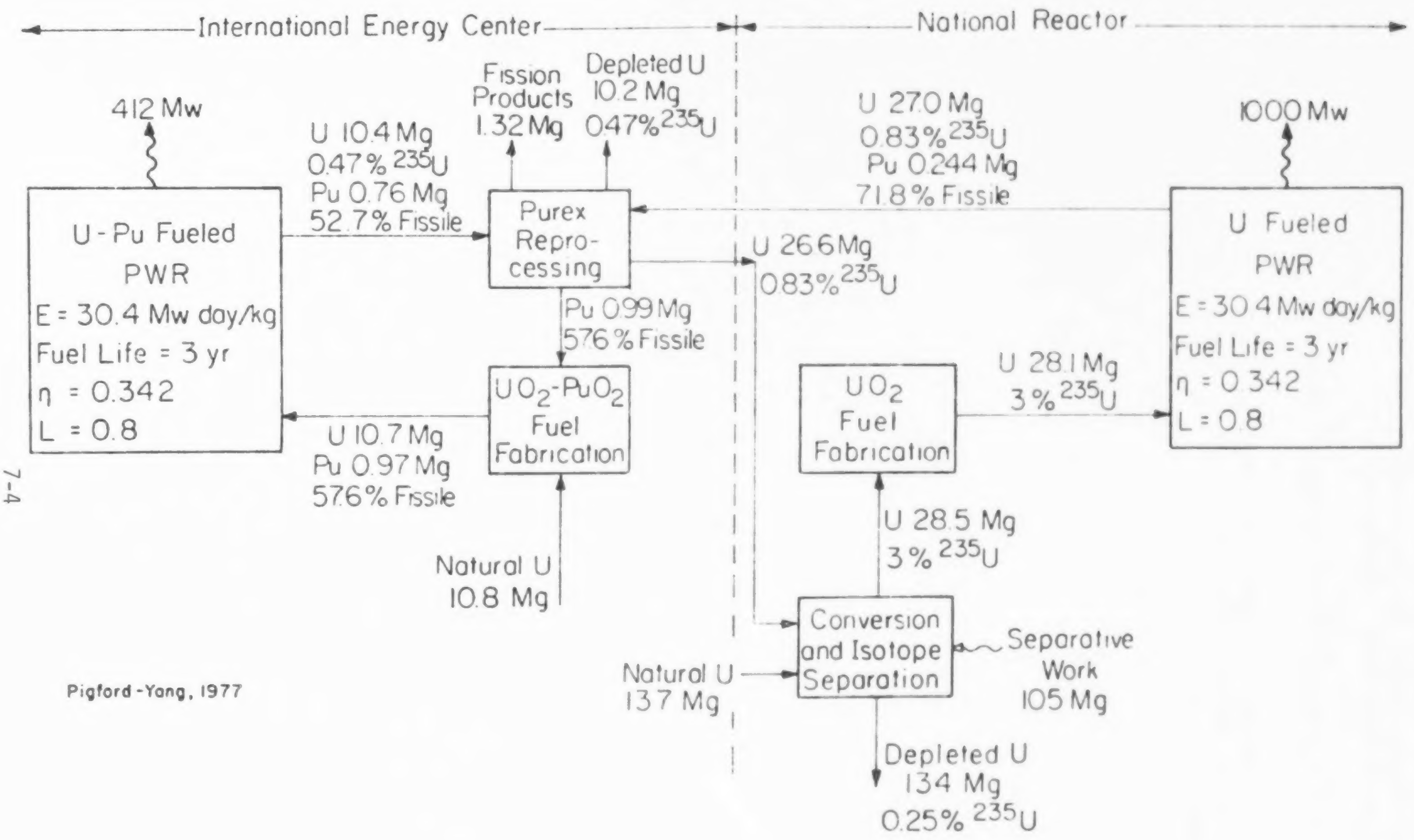
$$M_{238} \geq \left(\frac{1}{0.12} - 1\right) M_{233} + \left(\frac{1}{0.20} - 1\right) M_{235} \qquad (7.1)$$

Thorium is then added as additional fertile material so that the overall fissile concentration in the fuel is a few percent, typical of fuel for light-water reactors. This fresh fuel of denatured uranium and thorium is similar to low enrichment (i.e., "denatured") uranium fuel in that isotopic enrichment would be necessary in either case to produce uranium suitable for an explosive assembly. It differs in that much of the ^{238}U has been replaced by thorium, so that the production of chemically separable plutonium has been suppressed. However, appreciable quantities of plutonium are still present in the spent fuel, and the same set of issues as to its disposition still arise.

In Figure 7.2 is shown the overall equilibrium flowsheet for the international fuel cycle in which pressurized-water national reactors are fueled with denatured uranium and thorium, and plutonium is consumed in international plutonium-uranium-fueled pressurized-water reactors. The model reactor used in these calculations is that described in Section 3. The plutonium production per unit amount of ^{238}U in the national reactor fueled with uranium-thorium is 1.85 times greater than in uranium-fueled reactor of Figure 3.1. The lower concentration of ^{238}U in uranium-thorium fuel decreases the self shielding of the ^{238}U resonances, increases resonance absorption, and increases plutonium production per unit mass of ^{238}U in the fuel. Therefore, even though uranium-thorium PWR fuel contains 5.5 times less ^{238}U than the slightly enriched uranium fuel of Figure 3.1, the reduction in plutonium generation is not nearly so great.

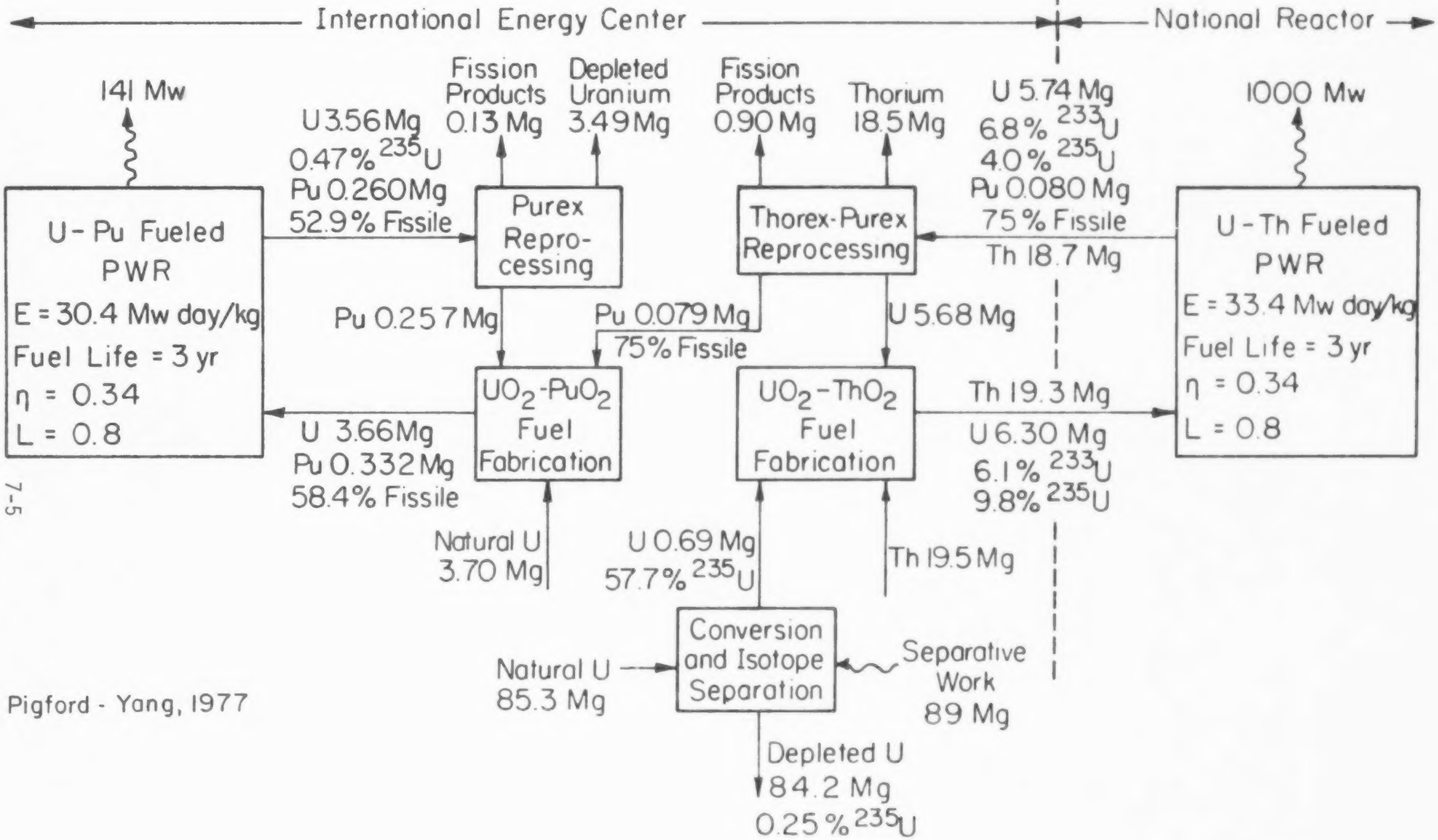
The discharge fuel from the national reactor could be shipped to an internationally controlled centralized reprocessing center. Although the plutonium could be allowed to follow the fission products to the high level wastes, the reprocessing chemistry is such that this would not materially simplify the separation operations. If the plutonium were to follow the high-level wastes, the fissile content of those wastes would be as high as 3 to 4 weight percent, which is much greater than that in discharge fuel from uranium-fueled water reactors. Thus, the high-level wastes would have to be safeguarded. Alternatively, the plutonium could be recovered and consumed in on-site plutonium-burner reactors, as shown in the overall flowsheet of Figure 7.2. Safeguards issues remain whether plutonium is allowed to follow the wastes or is consumed in a reactor at the international center. The effect of denatured-uranium thorium fueling is to reduce the necessary power of the international plutonium-burner reactor by a factor of 2.9 below that required with denatured uranium fueling.

As compared with the low-enrichment uranium cycle of Figure 7.2, the denatured uranium-thorium cycle has the advantage that a single international reprocessing center could service a larger number of national reactors, with only a relatively small total power of plutonium-burner reactors at the international center. However the required uranium enrichment capacity would be greater than in the case of the low-enrichment uranium cycle of Figure 7.1. The enriched product, containing about 58%


International Energy Center
National Reactor
412 Mw
U - Pu Fueled PWR
E = 30.4 Mw day/kg
Fuel Life = 3 yr
η = 0.342
L = 0.8
U 10.4 Mg
0.47% 235U
Pu 0.76 Mg
52.7% Fissile
Fission Products
1.32 Mg
Depleted U
10.2 Mg
0.47% 235U
Purex Reprocessing
Pu 0.99 Mg
57.6% Fissile
UO2-PuO2 Fuel Fabrication
U 10.7 Mg
Pu 0.97 Mg
57.6% Fissile
Natural U
10.8 Mg
U 27.0 Mg
0.83% 235U
Pu 0.244 Mg
71.8% Fissile
U 26.6 Mg
0.83% 235U
1000 Mw
U Fueled PWR
E = 30.4 Mw day/kg
Fuel Life = 3 yr
η = 0.342
L = 0.8
UO2 Fuel Fabrication
U 28.1 Mg
3% 235U
U 28.5 Mg
3% 235U
Conversion and Isotope Separation
Separative Work
105 Mg
Natural U
137 Mg
Depleted U
134 Mg
0.25% 235U

Pigford-Yang, 1977

Fig. 7.1 Annual quantities for LWR cycle for international safeguards, national reactors fueled with low enrichment (denatured) uranium. (E=fuel exposure, η=overall thermal efficiency, L=capacity factor)


International Energy Center
National Reactor
141 Mw
U-Pu Fueled PWR
E = 30.4 Mw day/kg
Fuel Life = 3 yr
η = 0.34
L = 0.8
U 3.56 Mg
0.47% ^{235}U
Pu 0.260 Mg
52.9% Fissile
Fission Products 0.13 Mg
Depleted Uranium 3.49 Mg
Purex Reprocessing
Pu 0.257 Mg
U 3.66 Mg
Pu 0.332 Mg
58.4% Fissile
UO_2-PuO_2 Fuel Fabrication
Natural U 3.70 Mg
Fission Products 0.90 Mg
Thorium 18.5 Mg
Thorex-Purex Reprocessing
Pu 0.079 Mg
75% Fissile
U 5.68 Mg
UO_2-ThO_2 Fuel Fabrication
U 0.69 Mg
57.7% ^{235}U
Th 19.5 Mg
Natural U 85.3 Mg
Conversion and Isotope Separation
Separative Work 89 Mg
Depleted U 84.2 Mg
0.25% ^{235}U
U 5.74 Mg
6.8% ^{233}U
4.0% ^{235}U
Pu 0.080 Mg
75% Fissile
Th 18.7 Mg
Th 19.3 Mg
U 6.30 Mg
6.1% ^{233}U
9.8% ^{235}U
1000 Mw
U-Th Fueled PWR
E = 33.4 Mw day/kg
Fuel Life = 3 yr
η = 0.34
L = 0.8

Pigford - Yang, 1977

Fig. 7.2 Annual quantities for LWR cycle for international safeguards, national reactors fueled with thorium and denatured uranium (E=fuel exposure, η=overall thermal efficiency, L=capacity factor)

^{235}U in uranium, would have to be safeguarded until it is diluted with the recycled uranium in the fuel fabrication facility. Also, the denatured U-Th cycle requires far more complicated reprocessing and fuel refabrication operations. The technology base for this fuel cycle would require further development and engineering scale-up before industrial-scale operations could begin.

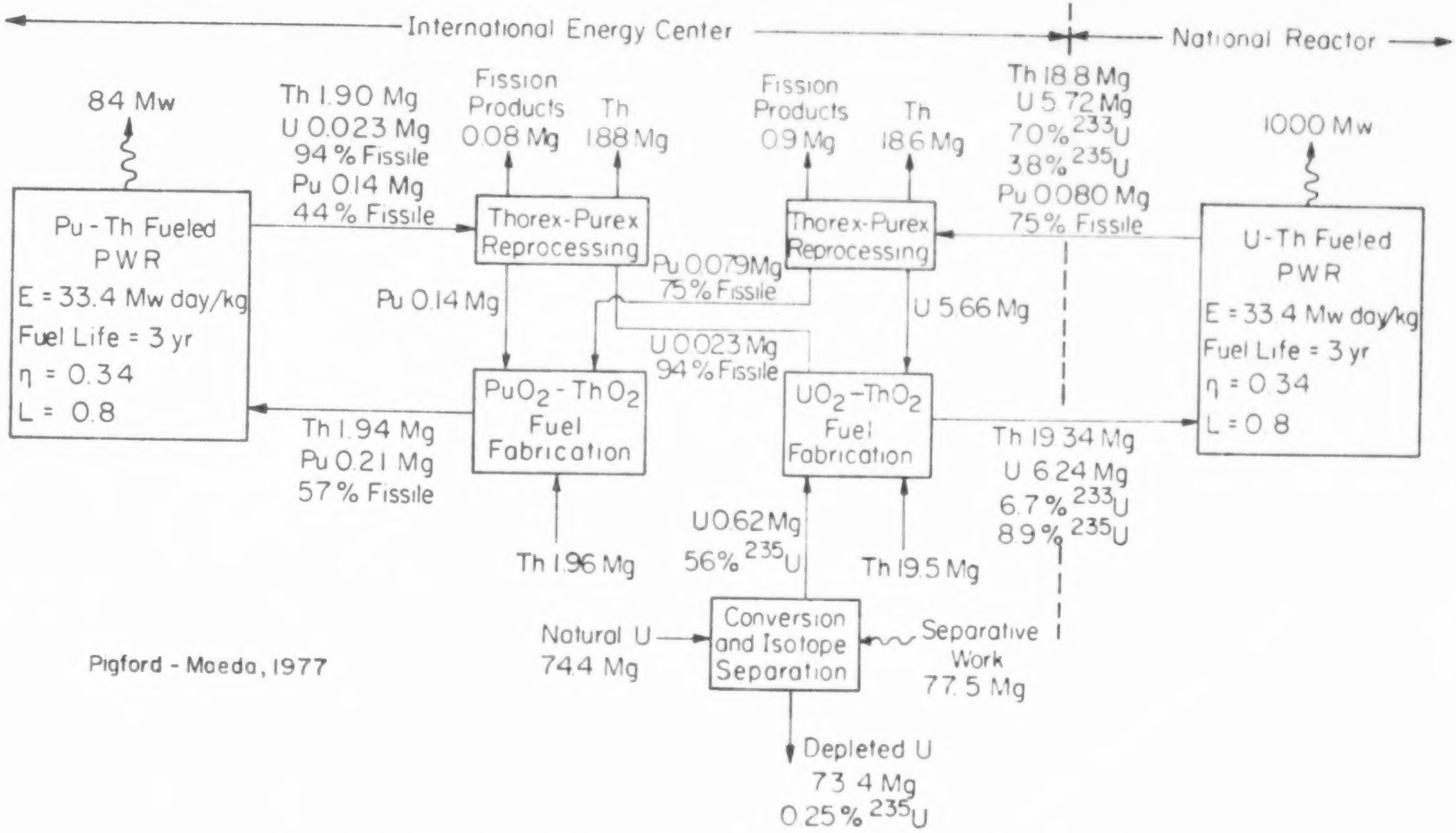
The flowsheets for equilibrium fuel cycles indicate less annual make-up uranium ore for uranium-thorium fueling. However, additional uranium ore is required to establish the equilibrium inventories in this fuel cycle. When evaluated on the basis of 30-yr lifetime uranium ore requirements for the same total system power, the uranium-thorium cycle of Figure 7.2 requires about 38% less uranium ore than the uranium cycle of Figure 7.1.

The relative power of the international plutonium-consuming reactor can be reduced further by using thorium instead of natural uranium as the fertile material for this reactor. The thorium is to be blended with plutonium from national reactors, as shown in Figure 7.3. Fissile uranium from thorium discharged from the international plutonium-burning reactor becomes an additional source of fissile make-up for the national denatured-uranium thorium reactors. Material quantities for the plutonium-thorium international reactor of Figure 7.3 were calculated from the data of Matzie (M2). This combination reduces the necessary power of the international plutonium-burning reactor by a factor of 4.9 below that of the simple uranium-plutonium cycles of Figure 7.1. The data shown here for the equilibrium fuel cycles indicate a further savings in the rate of consumption of uranium ore per unit of total electrical energy produced, as compared with Figure 7.2. However, the ore savings are not appreciable when accumulated over the reactor lifetime.

7.4 Denatured-Uranium-Thorium Cycle with National PWR and International LMFBR

Fast breeder reactors under international control could also be used as plutonium burners and as the source for the fissile uranium make-up for national denatured uranium-thorium reactors. Portions of the breeder blanket, such as part of the radial blanket, could contain thorium instead of depleted uranium. The thorium blanket would be reprocessed along with recycled uranium-thorium fuel from the national reactors. The recovered uranium would be diluted with natural or depleted uranium prior to off-site shipment as denatured uranium. It is likely that this concept could be technically possible by modifying the blanket loadings for even the first generation LMFBR's, which are expected to be started on plutonium.

A flowsheet of such a fuel cycle involving international breeder reactors is shown in Figure 7.4. This has been calculated from the characteristics of a commerical-scale LMFBR designed for possible introduction in this century (P2). It has been assumed that all of the breeding-gain fissile production of the breeder is drawn off as ^{233}U, to be used as fissile make-up for the denatured U-Th national reactors. As a result, no fissile breeding gain is available from this breeder to start up additional breeders, i.e., the effective doubling time for breeder fissile inventory becomes infinite. In principle additional breeder capacity could be introduced as needed, even when existing breeders operate at zero breeding gain, by starting the new breeders with isotopi-


International Energy Center
National Reactor
84 Mw
Pu - Th Fueled PWR
E = 33.4 Mw day/kg
Fuel Life = 3 yr
η = 0.34
L = 0.8
Th 1.90 Mg
U 0.023 Mg
94 % Fissile
Pu 0.14 Mg
44 % Fissile
Fission Products 0.08 Mg
Th 188 Mg
Thorex-Purex Reprocessing
Pu 0.14 Mg
PuO2 - ThO2 Fuel Fabrication
Th 1.94 Mg
Pu 0.21 Mg
57 % Fissile
Th 1.96 Mg
Pu 0.079 Mg
75 % Fissile
U 0.023 Mg
94 % Fissile
Fission Products 0.9 Mg
Th 18.6 Mg
Thorex-Purex Reprocessing
U 5.66 Mg
UO2 - ThO2 Fuel Fabrication
U 0.62 Mg
56 % 235U
Th 19.5 Mg
Natural U 74.4 Mg
Conversion and Isotope Separation
Separative Work 77.5 Mg
Depleted U 73.4 Mg
0.25 % 235U
Th 18.8 Mg
U 5.72 Mg
7.0 % 233U
3.8 % 235U
Pu 0.080 Mg
75 % Fissile
1000 Mw
U-Th Fueled PWR
E = 33.4 Mw day/kg
Fuel Life = 3 yr
η = 0.34
L = 0.8
Th 19.34 Mg
U 6.24 Mg
6.7 % 233U
8.9 % 235U

Pigford - Maeda, 1977

Fig. 7.3 Annual quantities for LWR cycle for international safeguards, national reactors fueled with thorium and denatured uranium, international reactors fueled with thorium and plutonium (E=fuel exposure, η=overall thermal efficiency, L=capacity factor)

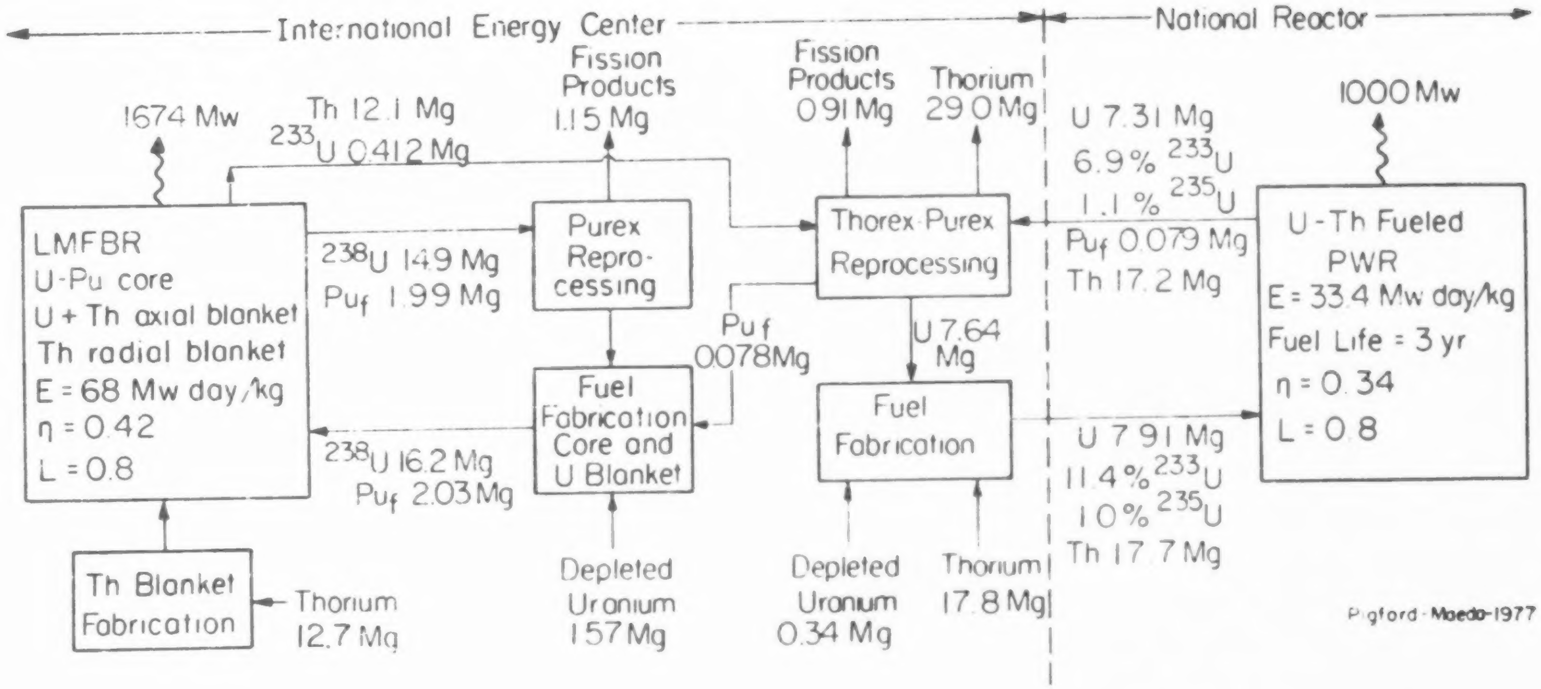

International Energy Center
National Reactor
1674 Mw
Th 12.1 Mg
^{233}U 0.412 Mg
Fission Products 1.15 Mg
Fission Products 0.91 Mg
Thorium 29.0 Mg
1000 Mw
LMFBR
U-Pu core
U + Th axial blanket
Th radial blanket
E = 68 Mw day/kg
η = 0.42
L = 0.8
^{238}U 14.9 Mg
Pu$_f$ 1.99 Mg
Purex Reprocessing
Thorex-Purex Reprocessing
U 7.31 Mg
6.9% ^{233}U
1.1% ^{235}U
Pu$_f$ 0.079 Mg
Th 17.2 Mg
U-Th Fueled PWR
E = 33.4 Mw day/kg
Fuel Life = 3 yr
η = 0.34
L = 0.8
Pu$_f$ 0.078 Mg
U 7.64 Mg
Fuel Fabrication Core and U Blanket
Fuel Fabrication
^{238}U 16.2 Mg
Pu$_f$ 2.03 Mg
U 7.91 Mg
11.4% ^{233}U
1.0% ^{235}U
Th 17.7 Mg
Th Blanket Fabrication
Thorium 12.7 Mg
Depleted Uranium 1.57 Mg
Depleted Uranium 0.34 Mg
Thorium 17.8 Mg
Pigford-Maeda-1977

Fig. 7.4 Annual quantities for national PWR fueled with thorium and denatured uranium, international LMFBR produces ^{233}U (E=fuel exposure, η=overall thermal efficiency, L=capacity factor)

cally enriched uranium. However, ^{235}U start-up is not an economical alternative for commercialized breeders (H1, P6). Therefore, operating an international breeder as shown in Figure 6.4 would be possible only after many decades when, even with an assumed zero growth of total fission electric power, the assumed breeders have finally been introduced to a level sufficient to replace the water reactors then being retired. Earlier operation would entail a much higher relative power from the international breeder, so that the breeder can then produce additional fissile material for start-up of new breeders.

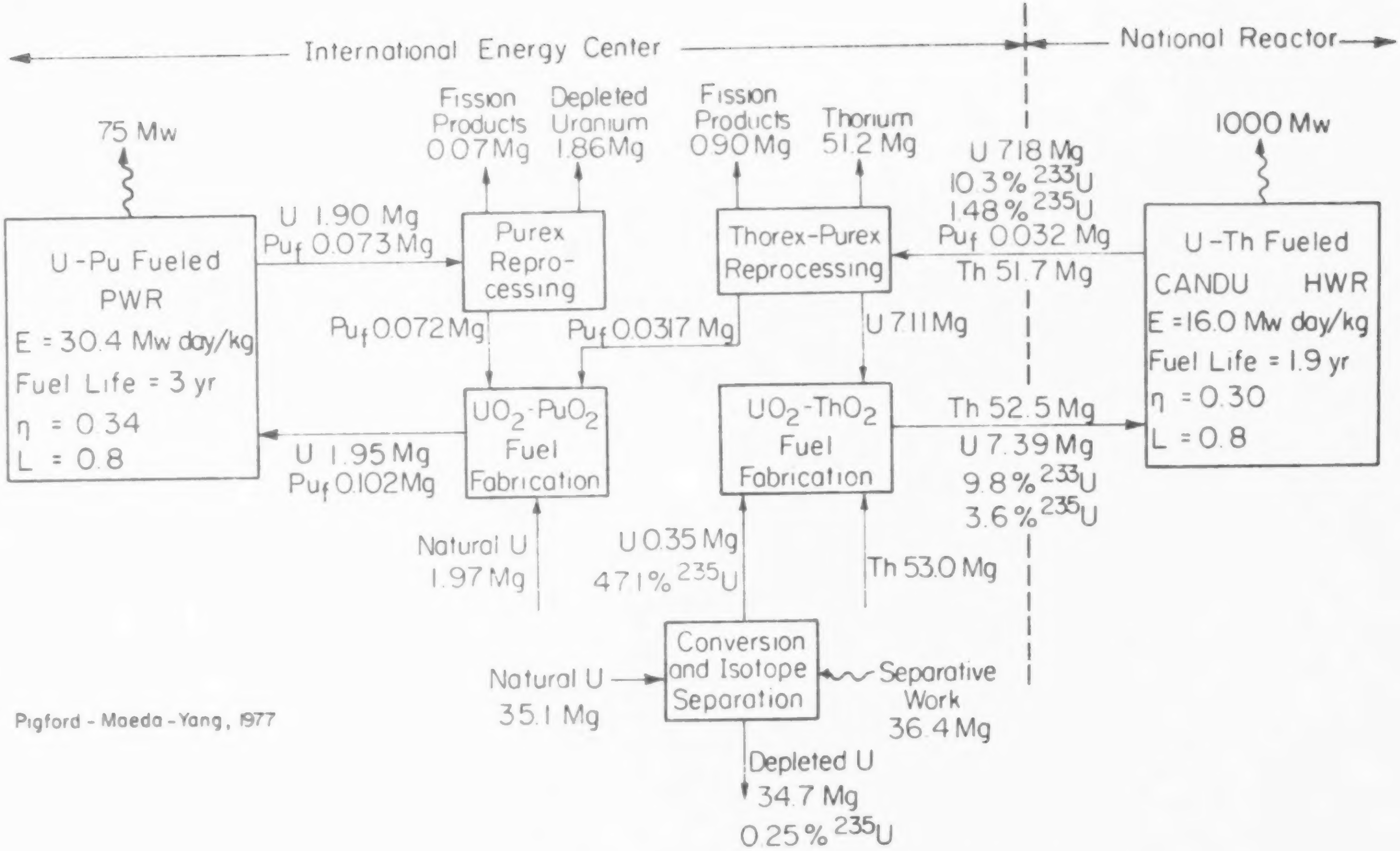
During the first few decades of breeder introduction the excess fissile production from these breeders is likely to be needed to start-up new breeders, so less fissile production is available as make-up for ^{233}U for national reactors. Consequently, the relative power of the international breeder shown in Figure 7.4 is the minimum breeder power, relative to the power of the national reactors, to supply the fissile make-up for the national reactors. To maintain a finite breeder doubling time an even larger relative power of the breeder would be required. However, the necessary breeder power can be reduced somewhat if the more favorable breeding gains calculated for future advanced breeders are assumed.

The fuel cycle flowsheet of Figure 7.4 has been calculated on the assumption that thorium can be used in both radial and axial blankets. The radial blanket alone will not produce sufficient ^{233}U at this power level. However, thorium in the axial blanket requires that the axial-blanket thorium pellets be segregated from the core pellets prior to reprocessing. Otherwise normal reprocessing of the entire fuel rod would dilute the ^{233}U with core uranium to the extent that it would be unsuitable for use in the denatured-uranium cycle. However, if the breeder power were increased to 3900 Mw, sufficient ^{233}U would be produced in the separate radial blanket, and normal head-end reprocessing techniques for core fuel could be used.

Fuel cycles involving denatured thermal uranium reactors and international breeders can provide excellent long-term ore utilization, but they require the greatest total power and the greatest reprocessing-refabrication capacity at the international facility. Also, such cycles have all the complexity of reprocessing and refabrication facilities necessary for both uranium-plutonium fueling and uranium-thorium fueling. They appear to be the least realistic in terms of time schedule and availability.

7.5 National and International Fast Breeders

It is also technically possible for the national reactor to be a breeder with a denatured ^{233}U-uranium core, with the fuel discharged from core and blanket sent to the international center for reprocessing. The breeder at the international center would consume the plutonium produced in the national breeder, and ^{233}U produced in the international breeder would be denatured by ^{238}U dilution and exchanged for the plutonium produced in the national breeder. However, full denaturing to 12% ^{233}U in uranium may not be possible because of the high fissile concentration in the breeder core required for criticality. Although the breeding gain possible with ^{233}U fueling in the breeder core is less than for plutonium


International Energy Center
National Reactor
75 Mw
U-Pu Fueled
PWR
E = 30.4 Mw day/kg
Fuel Life = 3 yr
η = 0.34
L = 0.8
U 1.90 Mg
Puf 0.073 Mg
Fission Products 0.07 Mg
Depleted Uranium 1.86 Mg
Purex Reprocessing
Puf 0.072 Mg
Puf 0.0317 Mg
UO2-PuO2 Fuel Fabrication
U 1.95 Mg
Puf 0.102 Mg
Natural U 1.97 Mg
Fission Products 0.90 Mg
Thorium 51.2 Mg
Thorex-Purex Reprocessing
U 7.11 Mg
UO2-ThO2 Fuel Fabrication
U 0.35 Mg
47.1% 235U
Th 53.0 Mg
Conversion and Isotope Separation
Natural U 35.1 Mg
Separative Work 36.4 Mg
Depleted U 34.7 Mg
0.25% 235U
U 7.18 Mg
10.3% 233U
1.48% 235U
Puf 0.032 Mg
Th 51.7 Mg
Th 52.5 Mg
U 7.39 Mg
9.8% 233U
3.6% 235U
1000 Mw
U-Th Fueled
CANDU HWR
E = 16.0 Mw day/kg
Fuel Life = 1.9 yr
η = 0.30
L = 0.8

Pigford - Maeda - Yang, 1977

Fig. 7.5 Annual quantities for national CANDU reactor fueled with thorium and denatured uranium, international Pu-burning PWR (E= fuel exposure, η=overall thermal efficiency, L=capacity factor)

in the core, there are possible safety benefits due to smaller changes in reactivity accompanying sodium voiding in the core. The international-national breeder system does provide less ore consumption than a system with thermal reactors at national sites, and thus it remains a possibility for the very long-term.

7.6 Denatured-Uranium-Thorium Stowaway Cycle for HTGR

To avoid the safeguards issues of normal thorium fueling with 93% ^{235}U make-up, as discussed in Section 7.1, the HTGR design can be adapted to denatured-uranium thorium fueling. The flowsheet for the near-equilibrium fuel cycle, without fuel reprocessing, has already been shown in Figure 5.3. Resource requirements are listed in Table 5.4.

7.7 Denatured Uranium-Thorium Cycles with National Heavy-Water Reactors

The pressure-tube heavy-water reactor, now commercialized in Canada as the CANDU reactor, is another possibility for a national reactor to be fueled with denatured uranium with or without thorium. The present cycle of the CANDU reactor fueled with natural uranium, with storage of discharge fuel, is the first possibility. Alternatively, this discharge fuel could be reprocessed at an international facility and the recovered plutonium could be consumed in an on-site plutonium-burner reactor. However, because of the expense of reprocessing fuel with the low concentration of plutonium formed in natural uranium fuel, the low burnup reprocessing cycle will not become economical until natural uranium prices become considerably higher than present contract prices. Instead, the national CANDU could operate with slightly enriched uranium for more economical reprocessing and more efficient resource utilization, qualitatively similar to the national reactor of Figure 7.1.

A CANDU national reactor could also be fueled with denatured uranium and thorium, as shown in Figure 7.5. The reactor lattice is assumed to be the same as in the present CANDU design. The fuel exposure of 16 Mwday/kg is that adopted in the study by Till and Chang (T1), and their data have been used to normalize the calculations for Figure 7.5. Because this fuel burnup is more than twice as great as that in the natural uranium CANDU, 9% void volume has been provided for fission gasses (T1). It is assumed, for the purpose of these calculations, that no other fuel modifications will be required. Even higher burnups would be expected for an optimum fuel cycle involving fuel with fissile concentrations greater than natural uranium. The national CANDU reactor of Figure 7.5 must be operated for many years with additional quantities of enriched ^{235}U before the equilibrium conditions shown in this figure are attained.

As compared with the cycle of Figure 7.2 involving the pressurized-water national reactor, the CANDU reactor of Figure 7.5 produces half as much fissile plutonium in the discharge fuel. It requires half the power of an international plutonium burner to consume the plutonium, or twice as many CANDU national reactors can be served by a single international plutonim-burner reactor. The lower plutonium generation of the denatured-uranium-thorium CANDU reactor is a consequence of the more moderated neutron spectrum and greater heterogeneity of the CANDU lattice.

Because of the low neutron absorption in deuterium and the lack of a pressure-vessel constraint in the CANDU reactor, sufficient heavy water is used as moderator so that the ratio of epithermal flux to thermal flux is much smaller than in the light-water reactor. This, together with the greater spatial self-shielding of resonance neutrons in the heavy-water lattice, results in less absorption in the ^{238}U resonances.. Because of the denaturing criterion of Equation (1), there are no large differences in the ratio of ^{238}U to fissile uranium in the PWR and CANDU reactors of Figures 7.2 and 7.5. In either reactor the rate of plutonium generation is proportional to the rate of absorption in ^{238}U divided by the fission rate. Therefore, because of the lower absorption in ^{238}U resonances in the CANDU, the plutonium generation in the U-Th CANDU is over two-fold less than in the U-Th PWR. The lower burnup in the CANDU does result in less plutonium consumption during irradiation than in the PWR, but the lower plutonium production rate is dominant and results in two-fold less plutonium in the U-Th CANDU discharge fuel.

The U-Th CANDU reactor of Figure 7.5 operates with an overall conversion ratio of 0.90, as compared with 0.67 for the U-Th PWR. Consequently more ^{233}U is bred in the CANDU, and much less fissile make-up and uranium ore are required. In both cases the make-up uranium is of sufficient enrichment that the enrichment supply and fuel fabrication must be under the same international safeguards.

Because of the radioactivity of 1.91-yr ^{228}Th and its daughters in irradiated thorium, the recovered thorium must be stored for several years before it can be recycled. Until such recycle occurs, the CANDU requires 2.7 times more make-up thorium than the U-Th PWR, because of the relatively low fuel exposure chosen for the CANDU and because of the higher concentration of thorium in the CANDU fuel.

The flowsheet for a national U-Th CANDU reactor fueled with make-up ^{233}U from an international breeder is shown in Figure 7.6. Even though the U-Th CANDU produces over two-fold less plutonium for the breeder fissile balance, it requires much less ^{233}U make-up because of its higher conversion ratio. Thus the required power of the international breeder for the U-Th CANDU cycle is 3.6 times smaller than that for the U-Th PWR cycle of Figure 7.2. The same considerations as to the time scale of feasibility of such a cycle, as discussed earlier for the U-Th PWR with an international breeder, also apply here.

These calculations indicate that when compared with a pressurized-water reactor, both operating on the denatured-uranium thorium fuel cycle, the CANDU heavy water reactor:

(a) produces about two-fold less plutonium,
(b) requires about two-fold less natural uranium for the make-up fuel and requires about two-fold less separative work if the make-up fissile uranium is obtained by isotope separation,
(c) requires about three-fold greater amount of thorium make-up fuel prior to thorium recycle, assuming the burnups used in this analysis,
(d) requires about two-fold less power of an international plutonium-burner reactor to consume the plutonium,

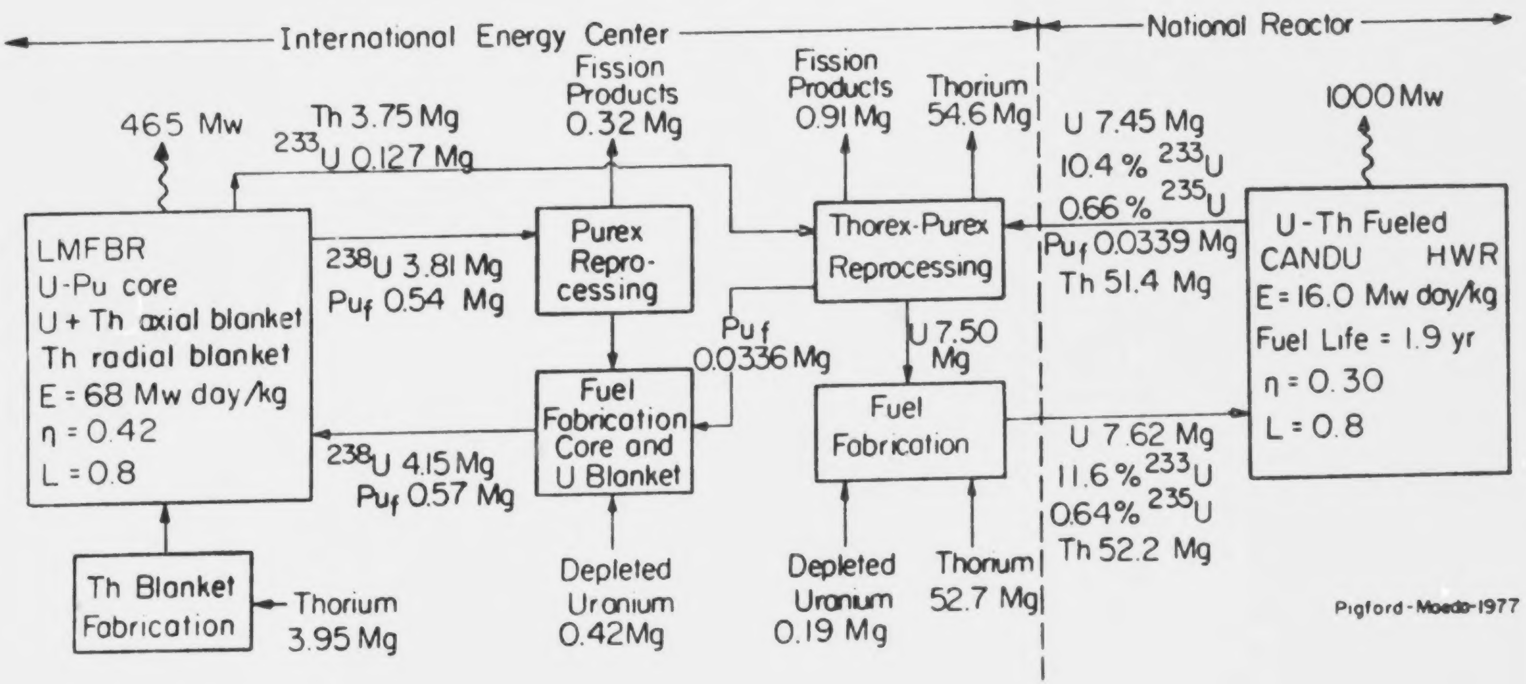

International Energy Center
National Reactor
465 Mw
Th 3.75 Mg
^{233}U 0.127 Mg
Fission Products 0.32 Mg
Fission Products 0.91 Mg
Thorium 54.6 Mg
1000 Mw
LMFBR
U-Pu core
U + Th axial blanket
Th radial blanket
E = 68 Mw day/kg
η = 0.42
L = 0.8
^{238}U 3.81 Mg
Pu$_f$ 0.54 Mg
Purex Reprocessing
Thorex-Purex Reprocessing
U 7.45 Mg
10.4 % ^{233}U
0.66 % ^{235}U
Pu$_f$ 0.0339 Mg
Th 51.4 Mg
U-Th Fueled CANDU HWR
E = 16.0 Mw day/kg
Fuel Life = 1.9 yr
η = 0.30
L = 0.8
Pu f 0.0336 Mg
U 7.50 Mg
Fuel Fabrication Core and U Blanket
Fuel Fabrication
^{238}U 4.15 Mg
Pu$_f$ 0.57 Mg
U 7.62 Mg
11.6 % ^{233}U
0.64 % ^{235}U
Th 52.2 Mg
Th Blanket Fabrication
Thorium 3.95 Mg
Depleted Uranium 0.42 Mg
Depleted Uranium 0.19 Mg
Thorium 52.7 Mg
Pigford-Maeda-1977

Fig. 7.6 Annual quantities for national CANDU reactor fueled with thorium and denatured uranium, international LMFBR produces make-up ^{233}U (E=fuel exposure, η=overall thermal efficiency, L=capacity factor)

(e) requires about four-fold less power from an international breeder if the fissile uranium make-up is obtained as excess ^{233}U from the breeder.

Our analysis is strictly limited to the considerations outlined herein, and there is no intent to imply conclusions as to the superiority of one reactor type over another. In further consideration of the CANDU reactor as a candidate for a safeguarded national reactor, it would be important to evaluate the required development of higher-burnup fuels for the CANDU, the licensability of the CANDU under the same criteria that are applied to light water reactors, the possible safeguards vulnerability of the CANDU because of its provision for frequent refueling with small fuel batches, and the relative costs of the CANDU and PWR systems.

Many of the features outlined above for the CANDU heavy water reactor as a national reactor fueled with denatured uranium would also apply to any other reactor of equivalent ratio, such as the more advanced high-conversion-ratio modifications of the HTGR reactor listed in Table 5.3. Light water reactors designed to high conversion ratio are also future possibilities (H1).

7.8 Enrichment Vulnerability of Denatured-Uranium Fuel

Using denatured uranium with a fissile content in the range of 10 to 20% creates a new safeguards issue in that relatively little work of isotope separation would be required to isotopically enrich this uranium to the level of highly enriched material. This can be illustrated in terms of the ^{235}U equivalent. Highly enriched uranium is usually regarded to be about 90% ^{235}U, which is made by isotopically enriching natural uranium. Of the total work required to enrich natural uranium to 90% ^{235}U, about 90% of the work is expended in enriching to 20% ^{235}U. Only 10% more work is required to further enrich to 90% ^{235}U. This illustrates the relative ease of making highly enriched uranium from uranium initially containing as much as 10 to 20% fissile concentration.

Because of the lower atomic mass of ^{233}U, the relative work required to enrich ^{233}U-^{238}U denatured uranium to the high-enrichment level would be even less than estimated above. Although recycled uranium containing ^{233}U and ^{232}U could not be enriched in commercial isotope separation plants because of the radioactivity, there are many relatively small and not necessarily efficient isotope separation systems that could enrich this uranium. The technology to carry out such enrichment on non-economical, non-commercial scale is available in the open literature. This is another aspect of the denatured uranium-thorium cycle that requires further evaluation.

To illustrate, the relative amounts of separative energy and plant capacity to produce 90% fissile uranium are estimated for the following two reactor fuels:

(a) normal PWR fuel, containing 3% ^{235}U in ^{235}U-^{238}U
(b) denatured uranium fuel containing 12% ^{233}U in ^{233}U-^{238}U.

For purposes of this illustration, we assume ideal close-separation cascades and 0.3% enrichment tails. The total interstage flow $\underline{J}$,per unit amount of product, is related to the ideal separation factor $\underline{\alpha}$ and to the separative work $\underline{Q}$ by (B6).

$$J = \frac{8Q}{(\alpha-1)^2} \qquad (7\text{-}2)$$

where

$$Q = \phi_p + \frac{x_p - x_F}{x_F - x_W}\,\phi_W - \frac{x_p - x_W}{x_F - x_W}\,\phi_F \qquad (7\text{-}3)$$

$$\phi = (2x-1)\,\ln\,\frac{x}{1-x} \qquad (7\text{-}4)$$

x = atom fraction of light isotope

and F, P, W denote feed, product, and tails, respectively. Also, for the close separation of heavy isotopes:

$$\alpha - 1 = \frac{A_i - A_j}{A_i + A_j} \qquad (7\text{-}5)$$

where A_i and A_j are the atomic weights of the heavy and light species, respectively. From these equations and assumed compositions, we calculate that

$$\frac{J \text{ for } {}^{235}\text{U}-{}^{238}\text{U}}{J \text{ for } {}^{233}\text{U}-{}^{238}\text{U}} = 7.8 \qquad (7\text{-}8)$$

Assuming that energy requirement and necessary equipment capacity are both proportional to the total interstage flow for a given method of separation (B6), about eight-fold less energy and plant capacity are required to enrich the ^{233}U-^{238}U fuel to 90% product than are required for the 3% ^{235}U-^{238}U fuel normally used in pressurized water reactors.

7.9 Comparison of Denatured-Uranium Fuel Cycles

The principal fuel cycle quantities for the various denatured fuel cycles are compared in Table 7.1. The uranium resource requirements, presented here in terms of the annual quantities of contained elemental uranium, are calculated for the equilibrium fuel cycles and do not reflect the additional non-equilibrium start-up requirements. If it is assumed that the international plutonium-burner reactor operates at a power level of 1000 Mwe, the last column represents the total electrical generating capability, in Gwe, of the system of national reactors and the international reactor which serves them. The annual resource requirements per unit of total system generating capacity are smallest when the international reactor is a breeder. Without breeders the smallest annual uranium requirement occurs for CANDU national reactors. The net thorium consumption by these reactors can be made small by later recycling the stored thorium.

A national denatured uranium-thorium PWR would reduce plutonium generation by only a factor of 2.9, and it could achieve a modest saving in uranium resources. Greater reductions in plutonium generation and in uranium-resource requirements are possible with a plutonium-thorium inter-

Table 7.1 Comparison of Fuel Cycle Quantities for Denatured Fuel Cycles

National Reactor Type/Fuel	International Pu-Burner Reactors Type/Fuel	Annual Resource Requirements[a] Mg/Gwe yr				Relative Electric Power	
		U	Th[c]	Separative Work	Th[d] Storage	National Reactors International Reactors	Total Systems International Reactors
PWR Denatured U (3% ^{235}U)	none	168.7[b]	-	108	-		
PWR Denatured U (3% ^{235}U)	PWR Pu + Natural U	104.7[b]	-	74.4	-	2.43	3.43
PWR Denatured U + Th	PWR Pu + Natural U	78.0[b]	17.1	78.0	16.2	7.09	8.09
PWR Denatured U + Th	PWR Pu + Th	68.6[b]	19.8	71.5	17.2	11.9	12.9
PWR Denatured U + Th	LMFBR Pu + depleted-U core Th + U axial blankets Th radial blanket	0.714[e]	11.4	-	10.8	0.60	1.60
PWR Denatured U + Th	LMFBR Pu + depleted-U core Th + U axial blankets Th radial blanket	0.815[e]	9.67	-	9.17	0.26	1.26
HTGR Denatured U + Th	none	136[b]	3.76	132	3.46		
CANDU Denatured U + Th	PWR Pu + Natural U	34.5[b]	49.3	33.9	47.6	13.3	14.3
CANDU Denatured U + Th	LMFBR Pu + depleted-U core Th + U axial blankets Th radial blanket	0.416[e]	38.7	-	37.3	2.15	3.15

[a] Calculated for equilibrium fuel cycle, total electrical generating capacity of system of international and national reactors = 1000 Mwe

[b] Natural uranium as U_3O_8 from milling and conversion of uranium ore

[c] Natural thorium (^{232}Th), as ThO_2 from milling and conversion of thorium ore

[d] Stored thorium can be recycled after storage for about 4 to 17 years

[e] Depleted uranium stockpiled from isotope separation

national reactor and with a national CANDU reactor fueled with denatured uranium and thorium.

It may be possible to turn to advantage the fact that all the denatured-uranium fuel cycles considered in this study require basically the same type of institutional and political agreements. In all cases the national reactors receive qualitatively similar denatured fresh fuel, and all national reactors discharge fuel containing unused energy resources, including enough plutonium to require that the discharged fuel be safeguarded. What changes from one cycle to another are the detailed facilities at the international sites. Therefore, if appropriate institutional and political agreements can be negotiated to make possible even the simplest of the cycles, i.e., the international stowaway fuel cycle, then substantially the same agreements and arrangements can remain in effect as more and more resource-efficient fuel cycles are introduced in the course of time. Thus it is important to fully analyze such safeguards fuel cycles for their economic, social and political consequences as well as their technical viability (U2).

8. Radioactivity, Long-Term Toxicity, and Actinide Content of High-Level Radioactive Wastes

8.1 Introduction

Here we present a comparison of the radioactivity and long-term toxicity of high-level radioactive wastes from the fuel cycles discussed in Chapters 3, 4, and 5. Emphasis is given to the actinides in the high-level wastes, which control the toxicity of these wastes after the fission product period of a few hundred years. For a more detailed comparison the transuranic wastes from fuel reprocessing and refabrication should also be considered, because the amount of actinide activity in these wastes is likely to be comparable to that in the high-level wastes (H1, P7, P8).

The waste toxicity considered here is the ingestion toxicity, defined as

$$\text{ingestion toxicity} = \sum_i \frac{\lambda_i N_i}{RCG_i} \qquad (8.1)$$

where N_i is the number of atoms of nuclide $\underline{i}$ in the wastes at any time $\underline{t}$, λ_i is the radioactive decay constant, and RCG_i is the radioactivity concentration guide for ingestion of nuclide $\underline{i}$, for unrestricted exposure to the public (U3).

It is to be emphasized that the ingestion toxicity of wastes, here presented on the basis of quantities per gigawatt-year of electrical energy generation, is only a crude and limited index of possible hazards of radioactive wastes. It does not take into account the long-term integrity of the waste form or the differences in transport of the different waste elements through the emplacement medium and through the environment.

8.2 Radioactive Wastes From the Reference U-Fueled Light-Water Reactor

The radioactivity of plutonium, americium, and curium in the high-level reprocessing wastes for the uranium-fueled water reactor of Figure 3.2 are shown in Figures 8.1 and 8.2 (P2). These quantities are calculated for the amount of wastes generated by reprocessing the fuel discharged yearly by a 1000 Mw reactor. The amount of ^{238}Pu in the high-level wastes increases with time because of the decay of ^{242m}Am and ^{242}Cm, the amount of ^{240}Pu increases because of the decay of ^{244}Cm and the amount ^{239}Pu increases because of the decay ^{243}Am and ^{243}Cm. The principal contributors to the long-term ingestion toxicity of these wastes are shown in Figure 8.3 (B1, H1, P1). During the first 600 years of waste storage the ingestion toxicity is dominated by ^{90}Sr in the fission products. Thereafter, ^{241}Am and ^{243}Am are most

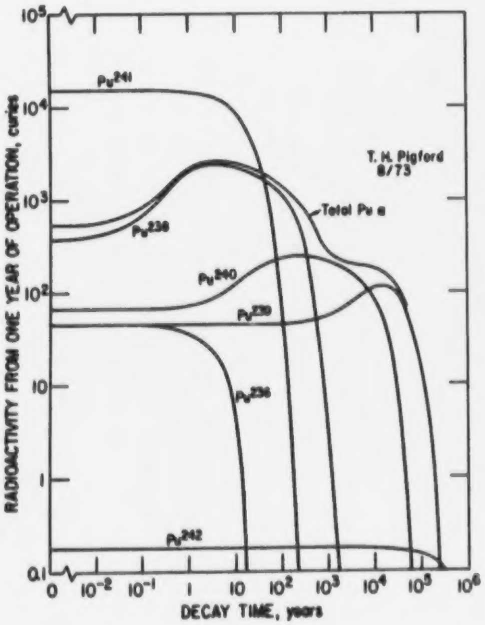


Fig. 8.1 Pu radioactivity in high-level wastes from U-fueled PWR (33 Mw day/kg, 0.5% U and Pu lost to waste)

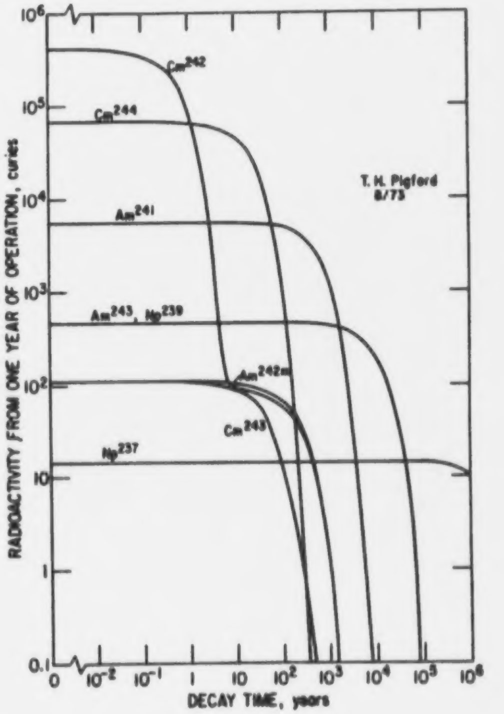


Fig. 8.2 Actinide radioactivity in high-level wastes from U-fueled PWR (33 Mw day/kg, 0.5% U and Pu lost to waste)

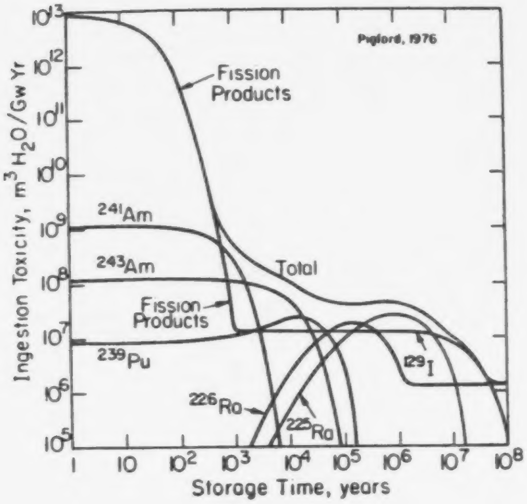


Fig. 8.3 Ingestion toxicity of high-level wastes from U-fueled PWR (33 Mw day/kg, 0.5% U and Pu lost to waste)

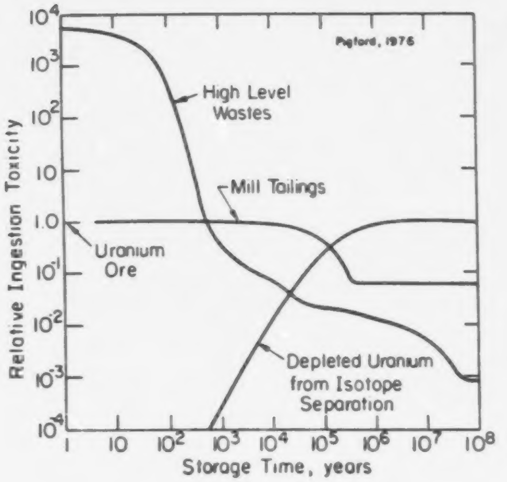


Fig. 8.4 Relative ingestion toxicity of fuel cycle residuals from U-fueled PWR (33 Mw day/kg, 0.5% U and Pu lost to waste, 5% lost to mill tailings, 0.25% ^{235}U in depleted uranium)

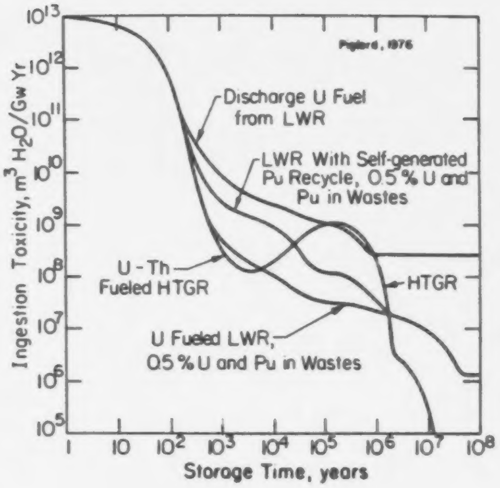


Fig. 8.5 Ingestion toxicities of high-level wastes from various fuel cycles (33 Mw day/kg for LWR's, 95 Mw day/kg for HTGR)

important, followed by ^{239}Pu and ^{240}Pu and then by ^{226}Ra and ^{225}Ra. The ^{226}Ra appears from the decay of ^{234}U, ^{238}Pu, ^{242m}Am, and ^{242}Cm initially in the high-level wastes, and ^{225}Ra is formed from the decay of ^{241}Pu, ^{241}Am, and ^{237}Np. After about 10^6 years of storage, the waste toxicity decays to a level due to ^{226}Ra in secular equilibrium with the small amount of ^{238}U in the high-level wastes. Although the long-lived fission product ^{129}I will be recovered separately from the bulk of the fission products containing the actinides, its long life and high toxicity require its inclusion in an overall toxicity analysis.

8.3 Waste Toxicities in Perspective

The ingestion toxicities for the high-level wastes from reprocessing non-recycled uranium fuel are compared with the toxicities of other residuals from this same fuel cycle in Figure 8.4 (B1, H4, P7). These toxicities are normalized to that of the uranium ore mined for one gigawatt year of reactor operation. The ore toxicity is due mainly to ^{226}Ra, which is in secular equilibrium in the ^{238}U decay chain. In the processes of milling and concentrating uranium ore ^{226}Ra and its precursor 80,000-yr ^{230}Th follow the tailings. Therefore, the ore toxicity is preserved in the mill tailings for a few hundred thousand years until ^{230}Th decays. Thereafter the tailing toxicity continues at a lower level determined by the residual uranium in the tailings, assumed here to be 5% of the uranium processes. If the depleted uranium from isotope separation is never used for breeder fuel, the uranium daughters, particularly ^{226}Ra, in this stored UF_6 will eventually be restored to a toxicity level with a few percent of the original ore toxicity.

The toxicity of the high-level wastes falls below that of the original ore after a period of about 600 years. The total toxicity of all residuals falls below that of the original uranium ore after a decay time of about 140,000 years. This minimum results from the enrichment of natural ^{234}U in isotope separation and its destruction in the reactor by neutron absorption, thereby depleting one of the sources of ^{226}Ra.

The toxicity indices are not measures of hazards, in part because they take no account of the barriers which isolate these wastes from the biosphere nor the behavior of different radioactive elements with respect to these barriers. However, the longer-term toxicities of the high-level reprocessing wastes are due to radium, which is the same element that controls the ore toxicity. The long-term radium toxicity of the reprocessing wastes is considerably less than the radium toxicity of the ore. It seems reasonable that radium ultimately appearing in the high-level wastes can be geologically isolated so that the waste material has less access to the environment than the radium in the natural ore.

A comparison of the hazards from high-level wastes and uranium ores can be derived from the results of Burkholder, et al (B8, B9), who have analyzed the long-term migration of fission products, actinides, and decay daughters from a model geologic repository; with sorption retardation of individual radionuclides according to chemical species. Hazards in terms of fifty-year integrated individual doses were calculated for migration times from 10^2 to 10^7 years after emplacement. Hazards from americium and plutonium were found to be less than the longer-term hazard from radium in the high-level waste. For migration pathways through the geologic medium as great as 480 meters the hazard from ^{226}Ra was found to be greater than the hazard from ^{90}Sr, the fission product which dominates the fission-product toxicity curve (Fig. 8.3) during the first few hundred years. The only fission products found to present greater hazards than ^{226}Ra were ^{99}Tc and ^{129}I, and then only for the longest geologic pathways and for relatively rapid leaching (0.3%/yr) of technetium and iodine from the wastes. Therefore, it is important to recognize that:

(a) the principal hazard from migration of radionuclides from high-level waste in geologic isolation may result from the long-term migration of ^{226}Ra, the same radionuclide that controls the ingestion and migration hazard from the original uranium ore and from the uranium mill tailings, and

(b) the amount of ^{226}Ra in the high-level wastes from reprocessing uranium fuel is less than the amount of ^{226}Ra in the ore mined to create these wastes (cf. Fig. 8.4).

Burkholder's (B8) analysis of hazards from radionuclide migration (B8), which assumes a ground water velocity as high as 110 meters/year, provides data on the effect of migration distance upon the 50-year dose from ^{226}Ra. Increasing the necessary migration distance from 160 meters, as might be representative of a shallow ore body, to as much as 16,000 meters, as might be obtainable in a geologic isolation, decreases the 50-year ^{226}Ra dose by a factor of twenty or more. Much larger attenuations occur for most other radionuclides. These ^{226}Ra doses are relatively insensitive to the dissolution rate of the radioactive source material, over a wide range of dissolution rates from 0.003 to 0.3%/yr.

These data for a model repository illustrate that high-level waste emplaced in a geologic repository, with sorption and transport properties representative of this model repository, may be expected to result in less actual hazard from nuclide migration than the hazards which would otherwise result from the ore body which produced these wastes; assuming that both of these sources of ^{226}Ra are exposed to the same mode of groundwater transport.

8.4 Effect of Pu Recycle on High-Level Waste Toxicity

Toxicities of high-level wastes from a light-water reactor with and without plutonium recycle are compared in Figure 8.5. Recycling plutonium increases the production of americium and curium (P2), whose radioactivity and decay daughters increase the ingestion toxicity by about an order of magnitude during the period governed by actinides and ^{226}Ra.

8.5 Toxicity of Unreprocessed Uranium Fuel

As shown in Figure 8.5, the actinide toxicity of unreprocessed uranium fuel from a light water reactor, which contains all of the plutonium discharged from the reactor, is about fifty times greater than the toxicity of wastes from uranium fuel which has been reprocessed for recovery of uranium and plutonium. This conclusion applies to the period from one thousand to one million years.

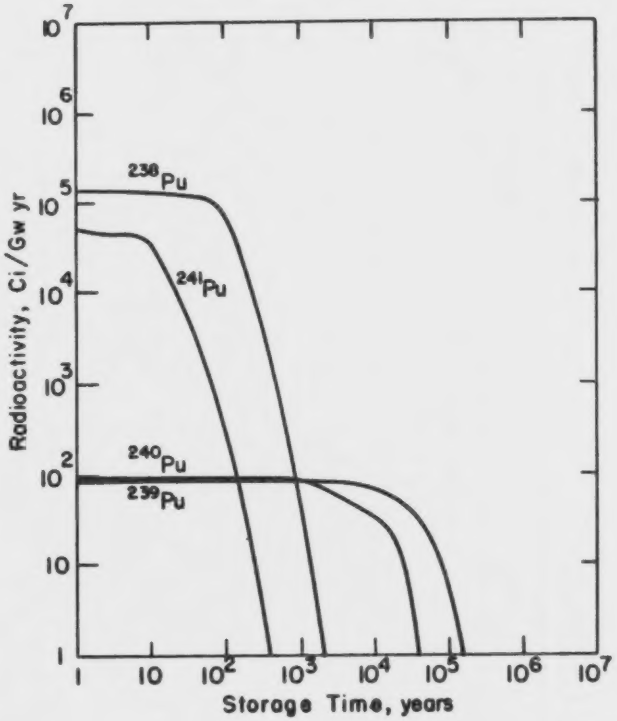


Fig. 8.6 Pu radioactivity in high-level wastes from ^{235}U-Th-fueled PWR with U recycle (33.4 Mw day/kg, 1.5% Th and U lost to waste)

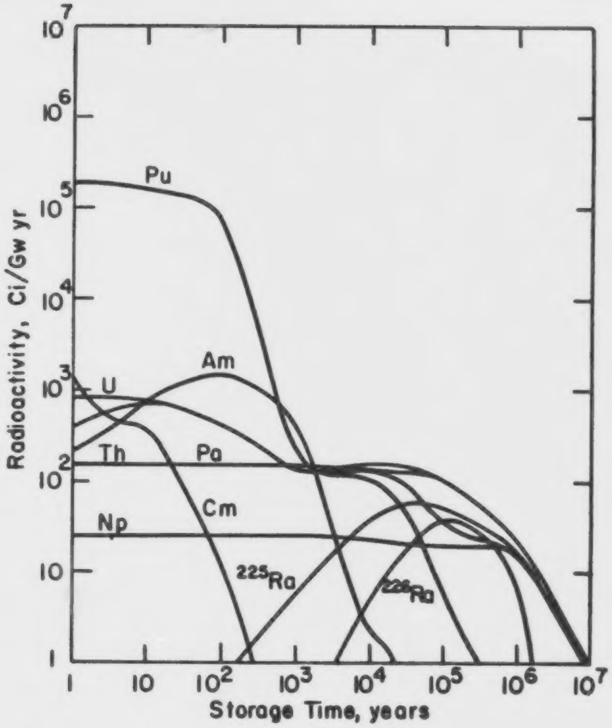


Fig. 8.7 Actinide radioactivity in high-level wastes from ^{235}U-Th-fueled PWR with U recycle (33.4 Mw day/kg, 1.5% Th and U lost to waste)

8.6 High Level Wastes from the PWR Fueled with ^{235}U, Th, and Recycled U

The radioactivity of plutonium radionuclides in the high-level reprocessing wastes for the equilibrium fuel cycle of the ^{235}U-Th fueled PWR are shown in Figure 8.6, calculated on the basis of all plutonium in the discharge fuel following the high-level wastes. The elemental radioactivity of the actinides and their daughters are shown in Figure 8.7. As compared with the plutonium in high-level wastes from reprocessing uranium fuel, the ^{235}U-Th PWR wastes contain over 100 times more ^{238}Pu, about the same quantities of ^{239}Pu, ^{240}Pu, and ^{237}Np, and 10^3 to 10^4 times 10^4 times less Am and Cm.

The ingestion toxicity of these thorium-cycle wastes is shown in Figure 8.8. Comparing with Figure 8.3 for the uranium-fuel wastes, the smaller amounts of Am and Cm in the thorium-cycle wastes result in relatively low waste toxicity after the fission-product period of about 600 years. The uranium activity and toxicity in these thorium-cycle wastes is relatively large because of the ^{232}U, ^{233}U, and ^{234}U in the recycled uranium, a fraction of which is lost to the wastes in each reprocessing cycle. The ^{234}U and ^{238}Pu result ultimately in the relatively large toxicity peak for ^{226}Ra.

The toxicity of the waste residuals from uranium and thorium milling for the U-Th-fueled PWR, as well as high-level reprocessing wastes, are plotted versus storage time in Figure 8.9. The presence of ^{230}Th in the natural thorium greatly increases the long-term toxicity of the milling residuals. At a concentration of 100 ppm of ^{230}Th, the ^{226}Ra daughter of ^{230}Th dominates the toxicity of the thorium ore. The ^{226}Ra remains with the tails from thorium milling, but it disappears by decay after about 10,000 years. Thereafter the toxicity of the thorium mill tailings reaches the level due to ^{230}Th-^{226}Ra in the residual thorium. A loss of 5% of the thorium to the tailings has been assumed.

With 100 ppm ^{230}Th in thorium ore, the toxicity of the thorium tailings is greater than that of the uranium tailings, both on the basis of fuel cycle quantities per unit of energy produced as well as on the basis of equal quantities of heavy element recovered. If the thorium is free of ^{230}Th, the thorium tailings have a lower ingestion toxicity than do the uranium tailings. The early toxicity of the tailings from pure ^{232}Th is due to 5.75-yr ^{228}Ra and its daughters. This decays after a few decades to the toxicity of the 5% residual ^{232}Th and its daughters in the tailings.

8.7 High Level Wastes from the PWR Fueled with Pu, Th, and Recycled U

Wastes from thorium fueling with plutonium make-up and uranium recycle include the high concentrations of ^{238}Pu, ^{232}U, ^{236}U, and ^{237}Np and their daughters resulting from the recycle of bred uranium as well as the high concentrations of americium and curium and their daughters resulting from plutonium irradiation. With plutonium make-up there is incentive to recover and recycle the plutonium remaining in the dishcarge fuel, and such recycle has been assumed in the fuel burnup calculations for this cycle. Therefore, for the purpose of waste calculations it is assumed that 1.5% of the plutonium and uranium in the discharge fuel is lost to the high-level wastes. The activities of plutonium radionuclides in the high level wastes from the equilibrium Pu-Th PWR fuel cycle are shown in Figure 8.10. The elemental activities are shown in Figure 8.11 and the ingestion toxicities are shown in Figure 8.12.

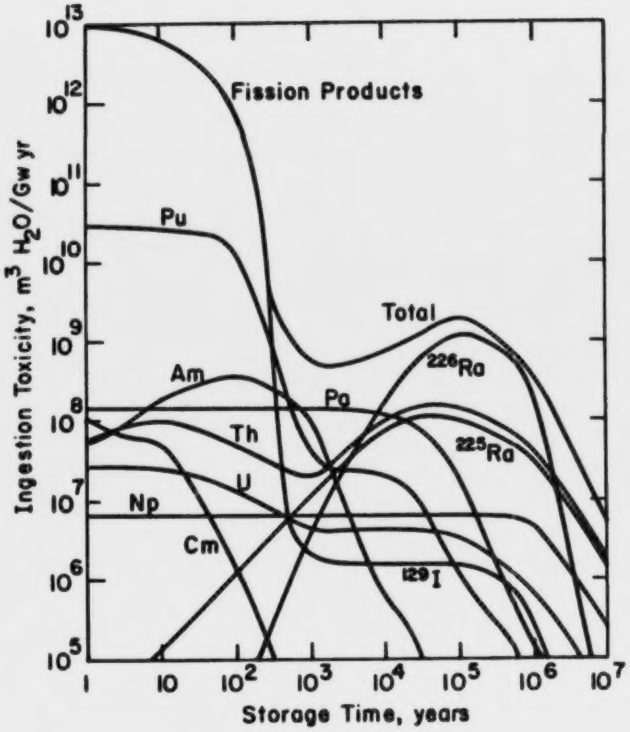


Fig. 8.8 Ingestion toxicity of high-level wastes from ^{235}U-Th-fueled PWR with U recycle (33.4 Mw day/kg, 1.5% Th and U lost to waste)

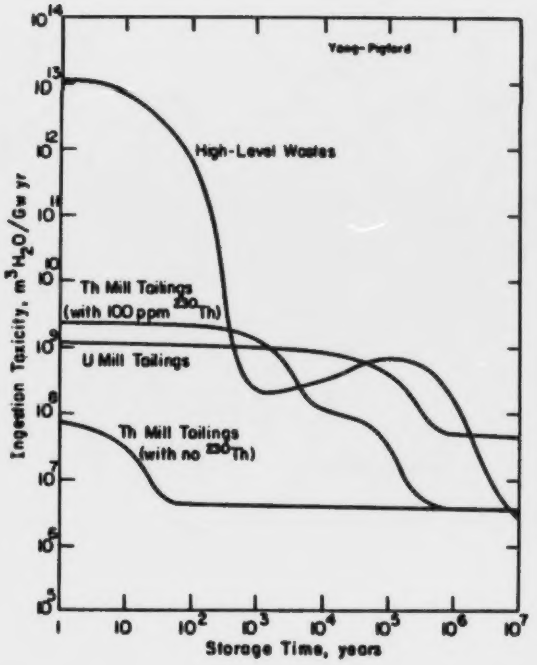


Fig. 8.9 Ingestion toxicity of fuel cycle residuals from ^{235}U-Th-fueled PWR with U recycle (33.4 Mw day/kg, 1.5% Th and U lost to waste, 5% lost to mill tailings)

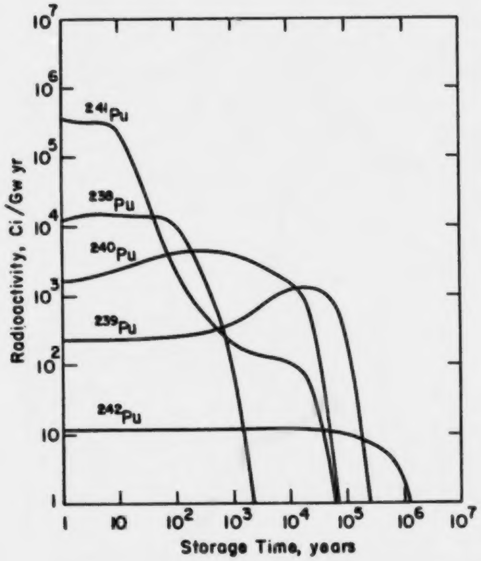


Figure 8.10 Pu radioactivity in high-level wastes from Pu-Th-fueled PWR with U and Pu recycle (33.4 Mw day/kg, 1.5% Th, U, and Pu lost to waste)

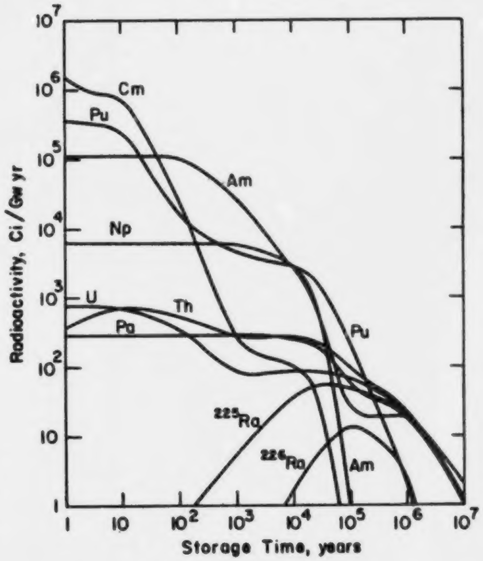


Fig. 8.11 Actinide radioactivity in high-level wastes from Pu-Th-fueled PWR with U and Pu recycle (33.4 Mw day/kg, 1.5% Th, U, and Pu lost to waste)

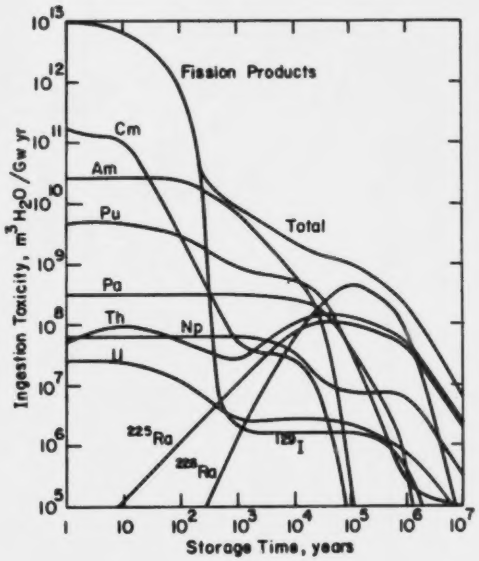


Fig. 8.12 Ingestion toxicity of high-level wastes from Pu-Th-fueled PWR with U and Pu recycle (33.4 Mw day/kg, 1.5% Th, U, and Pu lost to waste)

As a result of the increased americium, curium, and plutonium, the minimum in the waste toxicity at about 1000 years previously noted for ^{235}U-Th cycles does not occur. As shown in Figure 8.12, the effect of the plutonium make-up is to raise the waste toxicity by about sixty fold after the high-toxicity fission products have decayed. During the period of the ^{225,226}Ra peaks, the waste toxicity is essentially the same as for ^{235}U make-up.

The unusually high radioactivity of Np until about 100,000 years, as shown in Figure 8.11, is due to ^{239}Np in secular equilibrium with ^{243}Am. After Am decays the Np activity relaxes to the longer-term level due to ^{237}Np.

If all the plutonium in the discharge fuel were allowed to go directly to the high-level wastes there would be an increase by a factor of 67 in the initial activities of ^{239}Pu, ^{240}Pu, and ^{241}Pu, in Figures 8.10 and 8.11 and in the plutonium toxicity in Figure 8.12. However, the total waste toxicity is affected appreciably by plutonium only during a time interval at about 10,000 years of decay. At this time the important plutonium radionuclides are ^{239}Pu and ^{240}Pu. Most of the ^{239}Pu and much of the ^{240}Pu will have appeared from the decay of americium and curium in the wastes rather than from the plutonium initially in the wastes. Allowing all the plutonium to follow the wastes would not cause a significant increase in the total toxicity of these high-level wastes. Therefore, the main effect on waste toxicity resulting from choosing plutonium as fissile make-up in the Pu-Th-U cycle is the increased production of americium and curium.

8.8 High-Level Wastes from the Uranium-Fueled and Thorium-Fueled Heavy-Water CANDU Reactors

The actinide radioactivity and the ingestion toxicity of the discharge fuel from the uranium-fueled CANDU reactor are shown in Figures 8.13, 8.14, and 8.15. The activities of ^{239}Pu, ^{240}Pu, and ^{241}Pu are over 200-fold greater than in the high-level wastes from the uranium-fueled PWR (cf. Figure 8.1), because the CANDU fuel has not been reprocessed for plutonium recovery. The initial activity of americium in the CANDU fuel is about the same as in the U-PWR high-level wastes, but it increases about 10-fold in the first 100 years of storage, due to the decay of ^{241}Pu in the CANDU fuel. The total ingestion toxicity of the discharged CANDU fuel is comparable to that of unreprocessed PWR fuel shown in Figure 8.5.

The radioactivity and ingestion toxicities of high-level reprocessing wastes from the equilibrium fuel cycle of the ^{235}U-Th-fueled CANDU reactor, operating with uranium recycle, are shown in Figures 8.16, 8.17, and 8.18. The quantities are quite similar to

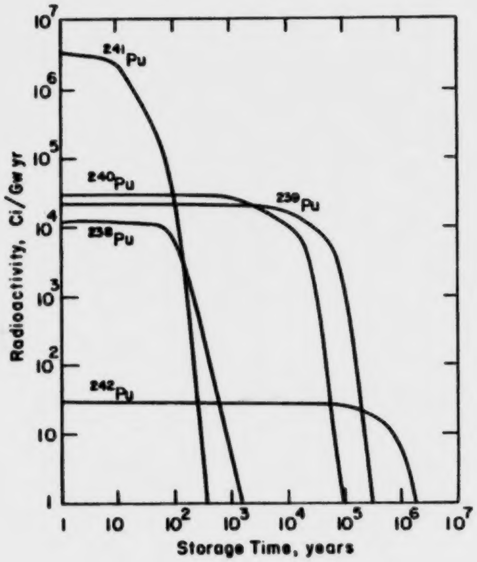


Fig. 8.13 Pu radioactivity in natural-U-fueled CANDU reactor discharge fuel (7.5 Mw day/kg)

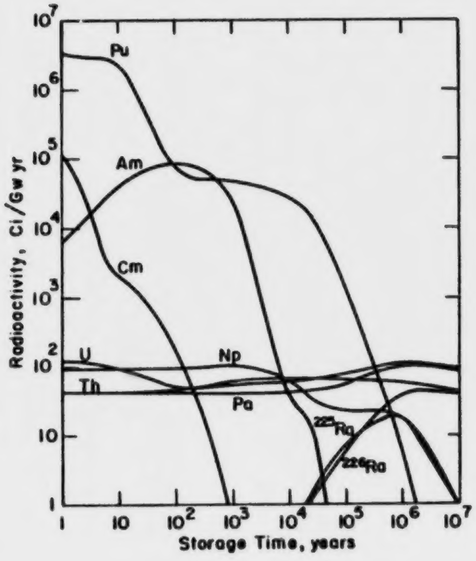


Fig. 8.14 Actinide radioactivity in natural-U-fueled CANDU reactor discharge fuel (7.5 Mw day/kg)

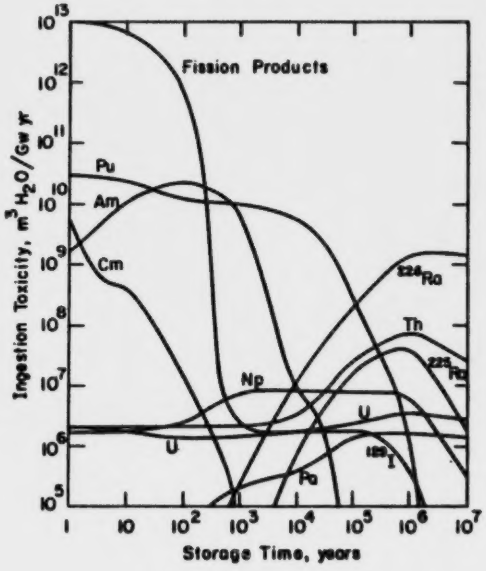


Fig. 8.15 Ingestion toxicity of natural-U-fueled CANDU reactor discharge fuel (7.5 Mw day/kg)

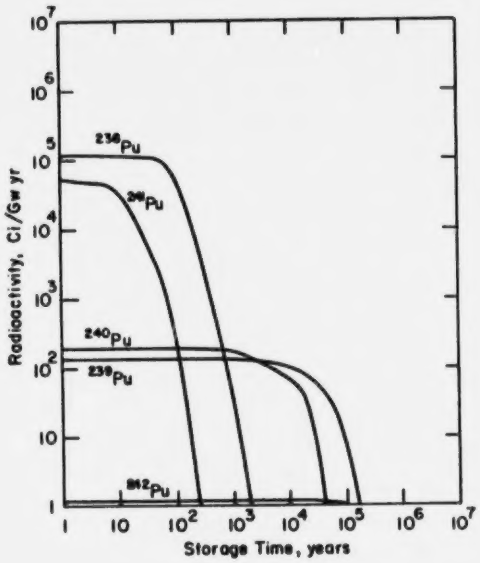


Fig. 8.16 Pu radioactivity in high-level wastes from ^{235}U-Th-fueled CANDU reactor with U recycle (27 Mw day/kg, 0.5% Th and U lost to waste)

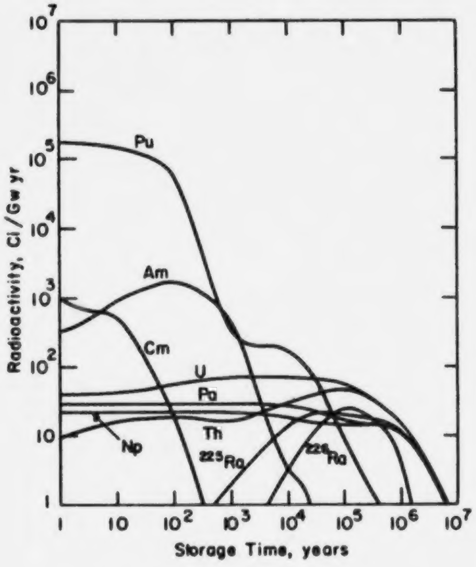


Fig. 8.17 Actinide radioactivity in high-level wastes from ^{235}U-Th-fueled CANDU reactor with U recycle (27 Mw day/kg, 0.5% Th and U lost to waste)

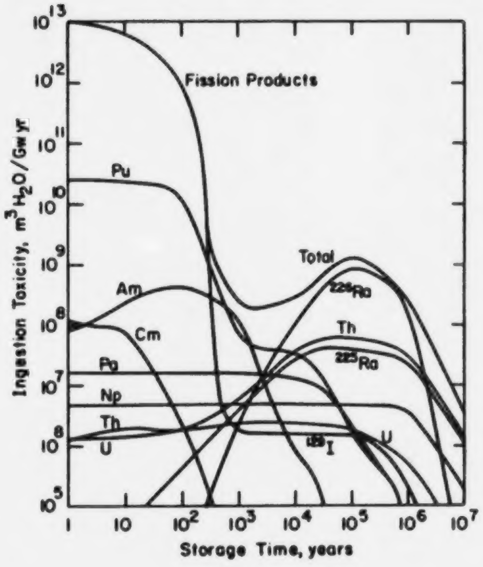


Fig. 8.18 Ingestion toxicity of high-level wastes from ^{235}U-Th-fueled CANDU reactor with U recycle (27 Mw day/kg, 0.5% Th and U lost to waste)

those in Figures 8.6, 8.7, and 8.8 for the ^{235}U-Th-fueled PWR. The peak amount of ^{225}Ra in the stored wastes is greater for the CANDU fuel cycle because of the greater throughput of ^{233}U in the reprocessing cycle, resulting in larger quantities of ^{233}U lost to the wastes.

The radioactivity and ingestion toxicities of high-level reprocessing wastes from the equilibrium fuel cycle of the Pu-Th-fueled CANDU, operating with U-Pu recycle, are shown in Figures 8.19, 8.20, and 8.21. All actinide quantities, except for ^{233}U, are smaller than in the case of the Pu-Th-fueled PWR.

8.9 High-Level Wastes from the Reference ^{235}U-Th-Fueled HTGR

The actinide radioactivity and ingestion toxicities of high-level reprocessing wastes from the equilibrium fuel cycle of the reference ^{235}U-Th-fueled HTGR, operating with uranium recycle and without reprocessing cross-over, are shown in Figures 8.22, 8.23, and 8.24. As compared with the waste properties for the ^{235}U-Th-fueled PWR, shown in Figures 8.6, 8.7, and 8.8, the HTGR wastes contain much greater radioactivity quantities of U, Np, Pu, Am, and Cm than do the PWR wastes. This is a consequence of the much higher burnup of the HTGR fuel cycle. Actinide cross-over in HTGR fuel reprocessing results in a small increase in the activities and toxicities of the HTGR wastes.

The total ingestion toxicity of the high-level wastes from the ^{235}U-Th HTGR is compared with that of other fuel cycles in Figure 8.5. The curve of the HTGR wastes is typical of that for any of the ^{235}U-Th fuel cycles. As has been explained in Section 8.6., the relatively small amounts of Am and Cm in ^{235}U-Th fuel cycle result in relatively little waste toxicity during the period of 10^3 to 10^5 years, after the fission products have decayed. However, the relatively large ^{226}Ra peak at 2×10^5 years for the HTGR ^{235}U-Th fuel cycle brings the toxicity of these wastes to the level of unreprocessed uranium fuel from PWR's.

Ingestion toxicities of long-term residuals from the HTGR ^{235}U-Th fuel cycle are shown in Figure 8.25. As compared with the similar plot (Figure 8.9) for the PWR ^{235}U-Th fuel cycle, the lower ingestion toxicity of the HTGR thorium mill tailings reflects the lower consumption of thorium in this fuel cycle, a consequence of the higher irradiation exposure of HTGR fuel. These differences in thorium consumption and thorium mill tailing toxicities become much smaller if thorium is recycled. The time trends of the toxicity of thorium mill tailings are explained in Section 8.6.

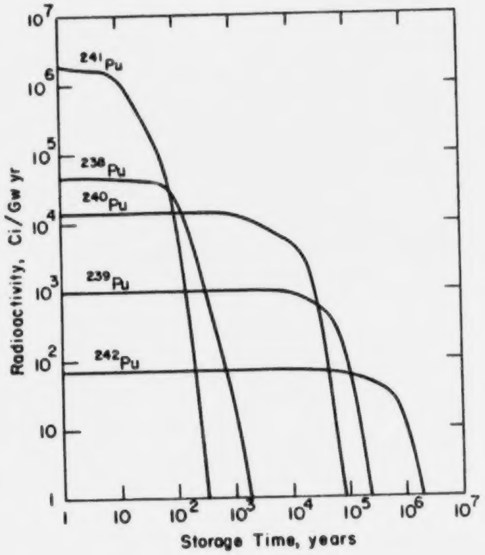


Fig. 8.19 Pu radioactivity in high-level wastes from Pu-Th-fueled CANDU reactor with U recycle (27 Mw day/kg, 0.5% Th and U lost to waste)

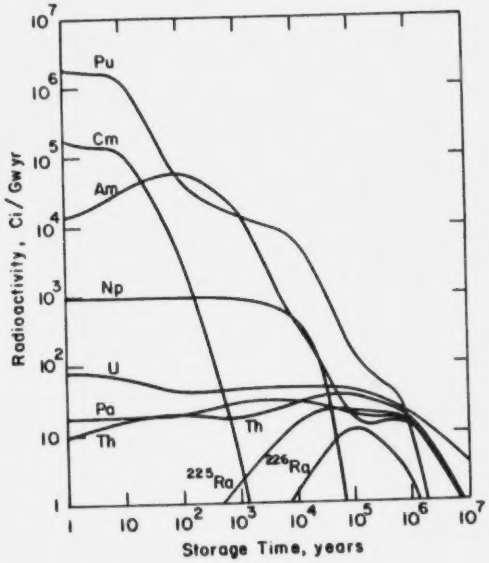


Fig. 8.20 Actinide radioactivity in high-level wastes from Pu-Th-fueled CANDU reactor with U recycle (27 Mw day/kg, 0.5% Th and U lost to waste)

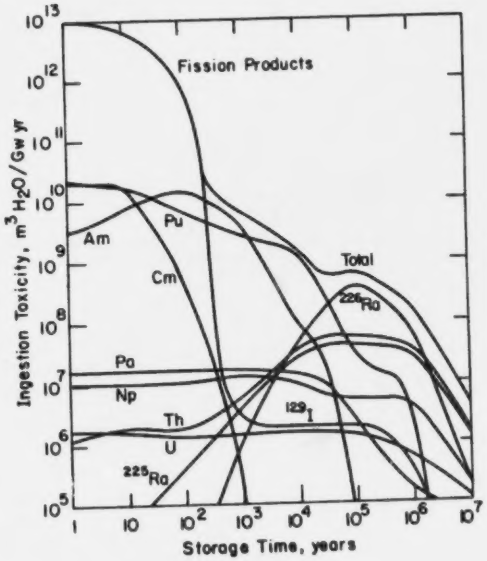


Fig. 8.21 Ingestion toxicity of high-level wastes from Pu-Th-fueled CANDU reactor with U recycle (27 Mw day/kg, 0.5% Th and U lost to waste)

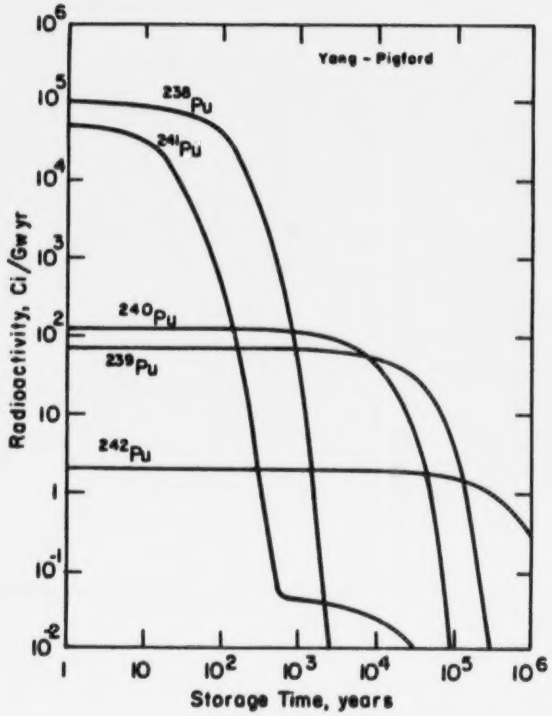


Fig. 8.22 Pu radioactivity in high-level wastes from ^{235}U-Th-fueled HTGR with U recycle (95 Mw day/kg, 0.75% U and Th lost to waste)

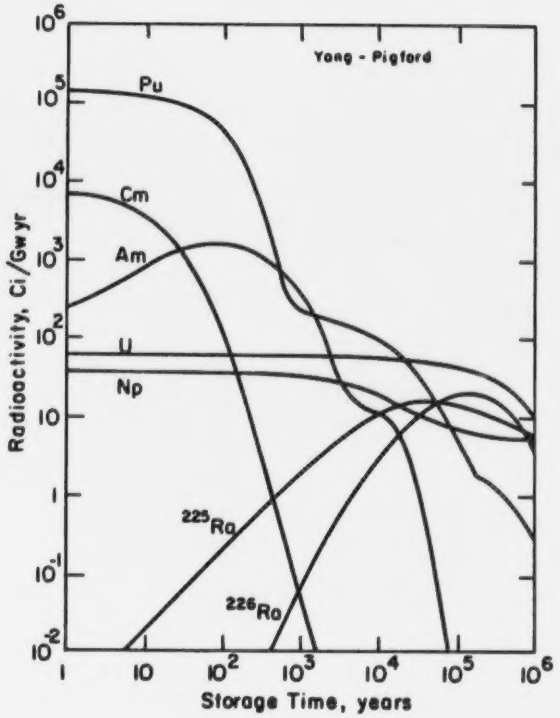


Fig. 8.23 Actinide radioactivity in high-level wastes from ^{235}U-Th-fueled HTGR with U recycle (95 Mw day/kg, 0.75% U and Th lost to waste)

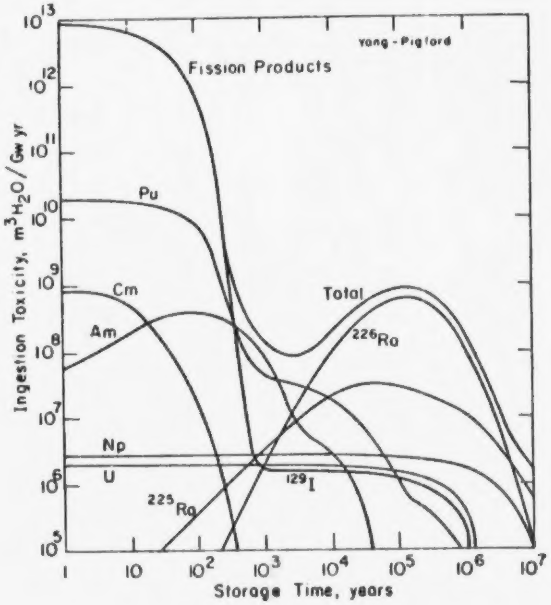


Fig. 8.24 Ingestion toxicity of high-level wastes from ^{235}U-Th-fueled HTGR with U recycle (95 Mw day/kg, 0.75% U and Th lost to waste)

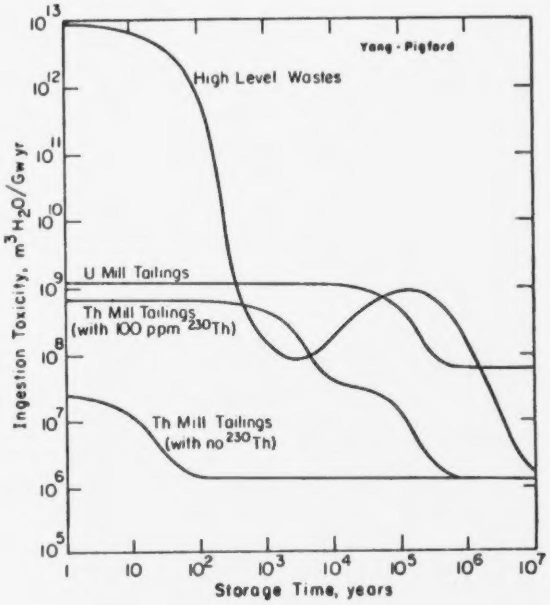


Fig. 8.25 Ingestion toxicity of fuel cycle residuals from ^{235}U-Th-fueled HTGR with U recycle (95 Mw day/kg, 0.75% U and Th lost to waste)

8.10 Comparison of Actinide Sources in High-Level Wastes From Alternate Fuel Cycles

The previous plots have shown that the hazard potential, i.e. the toxicity, of the high-level wastes is dominated by fission products for the first few hundred years, followed by ^{241}Am and ^{243}Am, then by ^{239}Pu and ^{240}Pu, and finally by ^{226}Ra and ^{225}Ra. The quantities of actinide precursors of each of these radionuclides are summarized in Table 8.1 for several alternate fuel cycles. For the purpose of comparison 0.5% of the recycled actinides are assumed to be lost to the high-level wastes.

Adopting the high level wastes from reprocessing uranium fuel from the pressurized-water reactor as a reference for comparison, the relative quantities of actinide precursors in the wastes from the other fuel cycles are characterized as follows:

1. The unreprocessed PWR discharge fuel will ultimately contain about 20 times more ^{241}Am, the same quantity of ^{243}Am, about 50 times more ^{239}Pu, ^{240}Pu, and ^{226}Ra, and about twice as much ^{225}Ra.

2. The high-level wastes from the U-Pu-fueled PWR with self-generated Pu recycle will ultimately contain about 4 times more ^{241}Am, 9 times more ^{243}Am, 7 times more ^{239}Pu, 14 times more ^{240}Pu 5 times more ^{226}Ra, and about the same quantity of ^{225}Ra.

3. The high-level wastes from the ^{235}U-Th-fueled PWR with self-generated uranium recycle will ultimately contain about 10 times less ^{241}Am, about 760 times less ^{243}Am, about 60% more ^{239}Pu, about 16% less ^{240}Pu, 42 times more ^{226}Ra, and about the same amount of ^{225}Ra.

4. The high-level wastes from the Pu-Th-fueled PWR, with U-Pu recycle, will ultimately contain about 23 times more ^{241}Am, 37 times more ^{243}Am, about 25 times more ^{239}Pu, ^{240}Pu, and ^{226}Ra, and about 80% more ^{225}Ra.

5. The high-level wastes from the ^{235}U-Th-fueled HTGR with self-generated uranium recycle will ultimately contain about 3 times less ^{241}Am, 16 times less ^{243}Am, half as much ^{239}Pu and ^{240}Pu, over 30 times more ^{226}Ra, and about half as much ^{225}Ra.

6. The high-level wastes from the U-Pu-fueled LMFBR will ultimately contain about 3 times more ^{241}Am, one-third less ^{243}Am, 3 times more ^{239}Pu, 2 times more ^{240}Pu, equal quantities of ^{226}Ra, and about half as much ^{225}Ra.

It has been concluded elsewhere (H1) that, in view of the anticipated efficacy of geologic isolation, the range of about 50 or less in the potential actinide hazards of the high-level wastes for the various fuel cycles considered herein does not appear to present a strong incentive for choosing one fuel cycle over another.

If it is assumed that the possible diversion and misuse of concentrated fissile material is to remain a long-term safeguards issue, as has been discussed in Chapter 7, then attention must be given to the long-term vulnerability of the appreciable quantities of fissile plutonium and fissile uranium in these radioactive wastes. The national-international safeguards fuel cycles discussed in Chapter 7 require the premise that, because of the intense radioactivity of fission products in discharge fuel, the plutonium in discharge fuel is sufficiently self-protected and suitable for storage at and shipment from dispersed national sites. Similar logic would apply to high-level wastes containing fissile actinides, as well as unreprocessed discharge fuel, during the first few hundred years of storage or disposal, while the fission products remain. Thereafter, the radioactivity of the wastes, per unit mass of plutonium contained in these wastes, is actually less than that of plutonium separated at the time of reprocessing. The ^{238}Pu and ^{241}Pu, which are the main contributors to plutonium radioactivity at the time of reprocessing, will have decayed away in these wastes after a few hundred years. Therefore, the fissile content of these wastes ultimately exists in a relatively non-radioactive environment.

Because of the radioactive decay of ^{243}Am, the amount of ^{239}Pu in the high-level reprocessing wastes increases with time, as has been demonstrated by the four-fold increase illustrated in Figure 8.1 for the reference high-level wastes and shown also in Table 8.1. Also, the fissile isotopic concentration in the plutonium present in these wastes, after the time period for americium decay, is greater than for plutonium recovered from discharge fuel.

The concentration of elemental plutonium in the reference uranium-fuel high-level reprocessing wastes, after americium decay, will be about 0.1 weight percent, assuming a four-fold dilution of the fission products and actinides by borosilicate glass. This compares with 0.94% for plutonium in discharged uranium PWR fuel. The chemical technology which can recover plutonium from discharge fuel in the presence of intense radioactivity can be reasonably expected to recover plutonium from the relatively nonradioactive high-level waste mixture after americium decay. Whether geologic isolation of these wastes for the purpose of environmental protection of future generations, and whether such safeguards issues for future generations are indeed relevant, are issues which may warrant further consideration.

Data in Table 8.1 show that the near-term inventory of fissile plutonium in unreprocessed discharge fuel is 200 times greater than that in the high-level reprocessing wastes at the time of reprocessing, and about 50 times greater when compared after storage long enough for americium decay. If short-term or long-term safeguards of fissile inventory in stored discharge fuel or in stored reprocessing wastes are important issues, then reprocessing to recover and consume the plutonium by recycle may be indicated (M5). However, the process of plutonium utilization increases the quantities of fissile plutonium ultimately in the wastes, as is illustrated in Table 8.1 for self-generated plutonium recycle in light water reactors and in fast breeders. The reduction ratio, i.e., the ratio of fissile plutonium inventory in discharge uranium fuel from PWR's to that ultimately appearing in high-level reprocessing wastes is about 10 for the recycle of plutonium in light-water reactors and is about 23 for utilization of plutonium to start first-generation fast breeders. The latter case is calculated on the basis of the breeder start-up requirements shown in Table 6.1 and the breeder waste inventories shown in Table 8.1. The reduction in plutonium inventory of wastes by recycling is much greater for the near term before americium decay. Data in Table 8.1 indicate a near-term reduction ratio of 140 for the PWR and 20 for the fast breeder.

If the fissile inventories in stored discharge fuel and in high-level reprocessing wastes are considered to be important short-term or long-term safeguards issues, then the non-reprocessing fuel cycle would clearly be the least favorable.

TABLE 8.1

Comparison of Actinide Quantities in High-Level Wastes from Alternate Fuel Cycles (Basis = 1 Gw yr of reactor operation, L = 0.8, 150 days preprocessing cooling, quantities calculated at time of reprocessing).

	U-fueled PWR a,b/	U-fueled PWR discharge fuel b/	U-Pu-fueled PWR self-generated Pu recycle a,b/	^{235}U-Th-fueled PWR U recycle c/	Pu-Th-fueled PWR U+Pu recycle a/	^{235}U-Th-fueled HTGR U recycle	U-Pu-fueled LMFBR a/
Sources of ^{241}Am, g atoms							
Pu - 241	5.75×10^{-1}	1.15×10^{2}	1.51	1.99	4.42	2.21	9.62×10^{-1}
Am - 241	5.47	5.47	2.49×10^{1}	1.54×10^{-1}	1.10×10^{2}	7.97×10^{-2}	1.68×10^{1}
Cm - 245	2.30×10^{-1}	2.30×10^{-1}	7.07×10^{-1}	1.17×10^{-3}	5.51	--	--
Total	6.22	1.21×10^{2}	2.71×10^{1}	2.15	1.20×10^{2}	2.29	1.78×10^{1}
Sources of ^{243}Am, g atoms							
Am - 243	1.02×10^{1}	1.02×10^{1}	8.97×10^{1}	1.30×10^{-1}	1.37×10^{2}	6.38×10^{-1}	7.90
Sources of ^{239}Pu, g atoms							
Pu - 239	3.01	6.02×10^{2}	4.29	6.05	5.21	4.94	3.05×10^{1}
Am - 243	1.02×10^{1}	1.02×10^{1}	8.97×10^{1}	1.30×10^{-1}	1.37×10^{2}	6.38×10^{-1}	7.90
Cm - 243	8.06×10^{-3}	8.06×10^{-3}	3.50×10^{-2}	--	--	5.35×10^{-4}	2.57×10^{-2}
Total	1.33×10^{1}	6.12×10^{2}	9.40×10^{1}	6.18	1.42×10^{2}	5.58	3.84×10^{1}
Sources of ^{240}Pu, g atoms							
Pu - 240	1.23	2.46×10^{2}	2.50	1.99	4.42	2.31	1.03×10^{1}
Cm - 244	3.73	3.73	6.39×10^{1}	2.53×10^{-2}	4.91×10^{1}	2.86×10^{-1}	0.520
Total	4.96	2.50×10^{2}	6.64×10^{1}	2.02	5.35×10^{1}	2.60	1.08×10^{1}
Sources of ^{226}Ra,d/ g atoms							
U - 234	6.71×10^{-2}	1.34×10^{1}	5.68×10^{-2}	5.05	3.68	3.30	2.88×10^{-3}
Pu - 238	1.26×10^{-1}	2.52×10^{1}	3.38×10^{-1}	3.36×10^{1}	1.79×10^{-1}	2.36×10^{1}	2.78×10^{-2}
Am - 242m	4.92×10^{-2}	4.92×10^{-2}	3.28×10^{-2}	1.22×10^{-3}	3.46	1.20×10^{-3}	2.94×10^{-1}
Cm - 242	5.50×10^{-1}	5.50×10^{-1}	2.95	4.95×10^{-3}	3.06	2.02×10^{-2}	4.68×10^{-1}
Total	7.92×10^{-1}	3.92×10^{1}	3.67	3.87×10^{1}	1.04×10^{1}	2.69×10^{1}	7.93×10^{-1}
Sources of ^{225}Ra, g atoms							
U - 233	--	--	--	9.58	8.96	4.06	--
Np - 237	8.61×10^{1}	8.61×10^{1}	6.37×10^{1}	1.17×10^{2}	1.33×10^{1}	4.60×10^{1}	2.16×10^{1}
Am - 241	5.47	5.47	2.49×10^{1}	1.54×10^{-1}	1.10×10^{2}	7.97×10^{-2}	1.69×10^{1}
Pu - 241	5.15×10^{-1}	1.15×10^{2}	1.51	1.99	4.42	2.21	9.62×10^{-1}
Cm - 245	2.30×10^{-1}	2.30×10^{-1}	7.07×10^{-1}	1.17×10^{-3}	5.51	--	--
Total	9.23×10^{1}	2.07×10^{2}	9.08×10^{1}	1.29×10^{2}	1.42×10^{2}	5.24×10^{1}	3.94×10^{1}

a/ High-level reprocessing wastes, 0.5% of U and Pu in discharge fuel appear in wastes. All other actinides in discharge fuel appear in wastes. For equilibrium fuel cycles.

b/ For PWR with 3.3% U fuel, E = 33 Mw day/kg, η = 0.325 (P2).

c/ High-level reprocessing wastes, 0.5% of U in first cycle ^{235}U and in bred U and 0.5% of Th appear in wastes. All other actinides in discharge fuel appear in wastes.

d/ Source which contribute to the ^{226}Ra peak of ∿190,000 yr. ^{238}U and ^{242}Pu are not included.

9. Generation of ^{14}C, ^{3}H, and other Radionuclides

9.1 Carbon-14

Carbon-14 is an activation product of potential environmental importance in the nuclear fuel cycle because of its long half life of 5,730 yr and because it easily appears in volatile form, such as CO_2. Most of the ^{14}C formed in reactors results from the (n,p) reaction with ^{14}N:

$$^{14}_{7}N + ^{1}_{0}n \longrightarrow ^{14}_{6}C + ^{1}_{1}H \qquad (9.1)$$

The ^{14}N, which constitutes 99.6% of natural nitrogen, is present as residual nitrogen impurity in oxide fuel of water reactors and fast-breeder reactors, as air dissolved in the coolant of water-cooled reactors, and as residual nitrogen in the graphite of high-temperature gas-cooled reactors. The ^{14}N activation cross section for 2200 m/sec neutrons is 1.85 barns.

Carbon-14 also results from the (n,γ) reaction on ^{17}O, which is present as 0.03% of natural oxygen, with a 2200 m/sec cross section of 0.235 barns:

$$^{17}_{8}O + ^{1}_{0}n \longrightarrow ^{14}_{6}C + ^{4}_{2}He \qquad (9.2)$$

In graphite-moderated reactors another source of ^{14}C is the (n,γ) reaction with ^{13}C, which is present as 1.108% of the natural carbon in graphite:

$$^{13}_{6}C + ^{1}_{0}n \longrightarrow ^{14}_{6}C + ^{0}_{0}\gamma \qquad (9.3)$$

However, the 2200 m/sec cross section is only about 0.9 millibarns. Additional but less important reactions are:

$$^{15}_{7}N + ^{1}_{0}n \longrightarrow ^{14}_{6}C + ^{2}_{1}H \qquad (9.4)$$

with a 2200 m/sec cross section of 2.4×10^{-7} barns, and

$$^{16}_{8}O + ^{1}_{0}n \longrightarrow ^{14}_{6}C + ^{3}_{2}He \qquad (9.5)$$

The activity $(N\lambda)_C$ of ^{14}C produced in a reactor can be estimated by assuming irradiation in a constant neutron flux for a period T_R. Because of the long half life of ^{14}C, the approximation $\lambda_C T_R << 1$ leads to

$$(N\lambda)_C = \lambda_C T_R \sum_i N_i \sigma_i \phi \tag{9.6}$$

where N_i = number of atoms of species i producing ^{14}C by neutron reactions

σ_i = cross section for species i to produce ^{14}C

λ_C = radioactive decay constant for ^{14}C.

Carbon-14 produced in water coolant is important because of its possible environmental release at the reactor site. If ^{14}C forms carbon dioxide or a hydrocarbon such as CH_4, and if no processes are provided to recover the gaseous ^{14}C, the coolant-produced ^{14}C will be discharged along with the non-condensable gases removed by the main condenser air ejector in a boiling water reactor and through the gaseous waste disposal system for a pressurized water reactor.

We consider here the production of ^{14}C by reactions (9.1) and (9.2) in the reactor coolant, which requires estimates of the inventories of ^{17}O and dissolved nitrogen in the coolant within the reactor core. For the 1000 Mwe PWR with an in-core water inventory of 13,400 kg, an effective $^{17}O(n,\alpha)$ thermal cross section of 0.149 barns, and an average thermal neutron flux of $3.5 \times 10^{13} n/cm^2sec$, the ^{14}C production from ^{17}O is estimated to be 2.2 Ci/yr.

To obtain the ^{14}C from dissolved nitrogen in the coolant, a dissolved nitrogen concentration of one part per million (by weight) is assumed, with an effective $^{14}N(n,p)$ cross section of 1.17 barns, resulting in a yearly production of 0.061 Ci of ^{14}C. The total yearly production of ^{14}C in the PWR coolant is then about 2.3 Ci/yr, which is the source term for possible environmental release at the reactor site. A 1000 Mwe boiling water reactor would contain about 33,000 kg of water in the core under operating conditions. Assuming the same values of neutron flux and cross sections, the yearly production of 5.6 curies of ^{14}C in the BWR coolant is estimated.

The ^{14}C produced by $^{17}O(n,\alpha)$ in UO_2 fuel, calculated as the yearly production per metric ton (Mg) of uranium originally in the make-up fuel, is

$$\frac{10^6 x 6.02 x 10^{23}}{238} \ \frac{\text{atoms U}}{\text{Mg U}} \times \frac{2x3.74x10^{-4} \text{ atoms } ^{17}O}{\text{atom U}} \times 6.47x10^{-25} \text{ cm}^2 \times$$

$$3.5x10^{13} \ \frac{1}{\text{cm}^2\text{sec}} \times \frac{\text{Ci}}{3.7x10^{10}\text{dis/sec}} \times \frac{\ell n2}{5730\text{yr}} \times 0.8 = 2.54x10^{-2} \text{ Ci/yr Mg-U}$$

For the ^{14}N source in the fuel, it is assumed that the nitrogen impurity is present in UO_2 at a weight ratio of 25 ppm, although nitrogen contents from 1 to 100 ppm have been reported (K2). The yearly production per metric ton of U is

$$10^6 \frac{\text{gram U}}{\text{Mg U}} \times \frac{270 \text{ gram UO}_2}{238 \text{ gram U}} \times 25\text{x}10^{-6} \frac{\text{gram N}}{\text{gram UO}_2} \times \frac{6.02\text{x}10^{23} \text{ atoms}}{14 \text{ grams}} \times$$

$$\frac{0.996 \text{ atoms } {}^{14}\text{N}}{\text{atom N}} \times 1.17\text{x}10^{-24} \text{ cm}^2 \times 3.5\text{x}10^{13} \frac{1}{\text{cm}^2\text{sec}} \times \frac{\text{Ci}}{3.7\text{x}10^{10}\text{dis/sec}}$$

$$\times \frac{\ln 2}{5730} \times 0.8 = 0.130 \text{ Ci/yr Mg U}$$

The total amount of ^{14}C produced yearly in the fuel is then 0.153 Ci per metric ton of uranium.

To obtain the ^{14}C in the discharge fuel, we use the fuel life of three calendar years for the reference pressurized-water reactor. Since there is negligible decay of the ^{14}C during this 3-yr period, the concentration in the discharge fuel is

$$3 \times 0.155 = 0.466 \text{ Ci/Mg}$$

The quantity of ^{14}C in the total fuel discharged yearly, which initially contained 27.2 Mg of uranium, is:

$$0.466 \times 27.2 = 12.7 \text{ Ci/yr.}$$

In a pressurized water reactor operating with plutonium recycle the thermal neutron flux is lower than for uranium fueling because of the higher fission cross section for plutonium. As a result, less ^{14}C is produced by thermal-neutron activation within the fuel, as shown in Table 9.1.

TABLE 9.1 ^{14}C in Discharge Fuel (1000 Mwe reactors, 80% capacity factor)

	PWR	PWR	HTGR	LMFBR
	U (3.3% ^{235}U)	U and recycled U + Pu	^{235}U, Th, and recycled U	U and recycled Pu
^{14}C, Ci/yr	12.7	6.7	120[a] 24[b]	3.3

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[a] Calculated for 30 ppm N_2 in HTGR graphite

[b] Calculated for 1 ppm N_2 in HTGR graphite

Fast-breeder oxide fuel is also assumed to contain 25 ppm of residual nitrogen(K2). Typical average fast-spectrum cross sections are 0.135 millibarns for $^{17}O(n,\gamma)$ and 14 millibarns for $^{14}N(n,p)$ within the reactor core (C3). For an average fast-spectrum core flux (C3) of $3.8\times10^{15} n/cm^2 sec$, and for the parameters of a near-term PuO_2 LMFBR (P2), the yearly production of ^{14}C for a 1000 Mwe fast breeder is estimated to be 3.3 Ci/yr. Relatively little ^{14}C is produced in the blanket fuel because of the lower neutron flux there.

The fuel of the high-temperature gas-cooled reactor (HTGR) consists of uranium and thorium particles, as oxides and carbides, distributed through a graphite matrix. The important ^{14}C-producing reactions in this fuel are $^{14}N(n,p)$ and $^{13}C(n,\gamma)$. Residual nitrogen is assumed to be present in graphite at a weight ratio of 30 ppm (B7). In the thermal-neutron energy spectrum of an HTGR the effective activation cross sections (B7)are 0.683 barns for ^{14}N and 3.3×10^{-4} barns for ^{13}C. For the average thermal-neutron flux of $1.2\times10^{14} n/cm^2 sec$ and a 4-yr fuel life, the estimated concentration of ^{14}C in the discharged graphite fuel is calculated from Eq. (9.6), with the result:

Source	Curies of ^{14}C per kg of graphite in discharge fuel
$^{14}N(n,p)$, 30ppm N	1.10×10^{-3}
$^{13}C(n,\gamma)$	2.29×10^{-4}
Total	1.33×10^{-3}

The fuel discharged yearly from the 1000 Mwe HTGR reactor of Fig. 5.1 contains 7.95 Mg of heavy metal and 90.5 Mg of graphite. The yearly production of ^{14}C by this reactor is then estimated to be

$$1.33\times10^{-3} \times 90{,}500 = 120 \text{ Ci/yr}$$

In other HTGR calculations 1 ppm of N_2 in the graphite is assumed (H4), resulting in an estimated yearly production of 24 Ci/yr for a 1000 Me plant.

When HTGR fuel is reprocessed the graphite matrix is to be incinerated in oxygen, exposing the fuel particles for dissolution. The combustion gas, which contains the ^{14}C and all of the normal carbon from the graphite, is to be recovered to avoid release of ^{14}C to the environment.*

The greater ^{14}C production in the HTGR, whether a factor of 2 or 10 times greater than in the oxide-fueled water reactors, is probably not the main issue in comparing the management of ^{14}C wastes in the HTGR cycle with ^{14}C management in LWR and CANDU cycles utilizing urania or thoria. When urania or thoria are reprocessed the ^{14}C released in fuel dissolution is diluted by normal carbon, in the form of CO_2 in the dissolver off-gas. An isotopic concentration of ^{14}C in CO_2 from dissolving oxide fuel of 130 to 650 ppm is estimated (D3). This is relatively concentrated when compared with the ^{14}C content of CO_2 released in HTGR fuel reprocessing, where very large quantities of normal carbon (90.5 Mg/yr) form CO_2 when the graphite fuel is incinerated. For 30 ppm N in the graphite, the resulting isotopic concentration of ^{14}C in carbon is 0.3 ppm, and it decreases to 0.06 ppm for 1 ppm N_2 in the graphite. This large volume of CO_2 , containing relatively small concentrations of radioactive gases, creates a challenging problem for fuel reprocessing development. Because the CO_2 interferes with the processes normally used to concentrate and remove ^{85}Kr from air streams, a new krypton-removal process is under development for HTGR fuel reprocessing.

In the HTGR reprocessing, the incinerator gases contain considerable carbon monoxide, so the filtered gas is first passed over a catalyst to oxidize CO to CO_2. Also, that portion of the tritium which may be in the form of HT is oxidized to HTO. Elemental radiodine is removed from the CO_2 by adsorption on a bed of lead zeolite, followed by a bed of silver zeolite for final elemental iodine cleanup and to remove methyl iodide. Tritiated water is removed on molecular sieves. Because of the low concentration of HTO in the CO_2 gas, it may be necessary to inject steam or water vapor upstream of the adsorbent bed as a carrier for HTO removal. After removal of ^{220}Rn and ^{85}Kr, CO_2 is finally reacted with a lime solution to precipitate calcium carbonate, which is filtered, dried, and packaged for disposal as low-level waste.

Incineration of HTGR discharge fuel from one gigawatt-year of operation results in the production of 332,000 kg of CO_2. This forms 754,000 kg of $CaCO_3$, which contains ^{14}C at the activity level of 32 to 160 nanocuries per gram.

Although ^{14}C is not an alpha emitter, its long life and its importance as a source of whole-body and organ dose suggest that these large quantities of $CaCO_3$ containing ^{14}C may have to be treated in a manner similar to alpha-contaminated wastes. A threshold of 10 to 100 nanocuries per gram for alpha-contaminated wastes and the practicalities of monitoring at this level would evidently dictate that essentially all of the reprocessing wastes must be treated as TRU wastes, to be disposed of ultimately in a geologic repository. The alpha contamination of the $CaCO_3$ is unknown. Even if the $CaCO_3$ is not alpha contaminated, the logic of the 10 to 100 nanocurie/gram threshold may apply as well to these ^{14}C wastes. If this were to result in a requirement that the HTGR-produced $CaCO_3$ be emplaced in the federal geologic repository, along with TRU wastes, a substantial penalty could accrue to the HTGR fuel cycle.

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*Normal atmospheric carbon already contains ^{14}C, produced by interaction of cosmic rays with nitrogen in the upper atmosphere. This natural concentration which existed prior to the atmospheric testing of nuclear weapons was 6.08×10^{-9} Ci per kilogram of atmospheric carbon, which is about 2×10^{5} less than the calculated concentration of ^{14}C in the carbon in HTGR discharge fuel.

Croff(C4) has analyzed the cost of various alternative means of managing the $CaCO_3$ produced in HTGR reprocessing. Burial in a geologic repository was found to be the most expensive of the alternatives considered, with an estimated cost in constant 1975 dollars of $280 per kilogram of heavy metal (Th + U) reprocessed. Because of the high burnup of the HTGR cycle, the economics of this fuel cycle are less affected by high unit costs of fuel cycle operation than in the case of the thoria fuel cycles. However, Croff's estimate for $CaCO_3$ disposal is over five-fold greater than the total cost, in the same constant dollars, estimated for off-site disposal of all wastes from reprocessing urania fuel (U4). Assuming generally similar wastes and waste costs from thoria fuel as from urania fuel, the additional costs of disposing of the large quantities of ^{14}C-contaminated $CaCO_3$ may impose a significant economic penality on the HTGR fuel cycle.

The issue of ^{14}C and how it must be disposed of also illustrates the problem of developing adequate and meaningful criteria for long-term waste management. As compared to the total actinides in reprocessing wastes, ^{14}C contributes little to the total activity. Its contribution to total ingestion toxicity is even lower, because its RCG is several orders of magnitude less than that of the actinides. When using the calculated waste toxicity as a criterion of hazards, ^{14}C would be considered to be relatively unimportant.

However, an evaluation of the hazards from waste disposal must also take into account the mechanisms and probabilities of the radionuclides reaching the biosphere. As an example of such an approach, Burkholder (B8, B9) has calculated the migration of radionuclides through a geologic medium. He assumed that all radionuclides in the geologic repository are leached into ground water at the same rate. The different equilibria between the various diffusing species and the soil through which they migrate were taken into account, and a constant linear velocity of ground water was assumed. The calculated amounts of radionuclides which appear at various distances and at various times from the position of emplacement indicate that, for the desert-soil properties and other conditions assumed by Burkholder, ^{14}C delivers a greater whole body and organ dose than any of the other radionuclides in the reprocessing wastes, which reflects the relatively high mobility of carbon compounds in such geologic media. Burkholder's ground-water model is highly simplified, and the parameters assumed in his calculations may not be appropriate for waste forms and geologic media that are finally selected. However, his calculations do illustrate the kind of methodology that should be developed, and they suggest that it is premature to draw conclusions on the importance of geologic isolation of ^{14}C on the basis of the calculated toxicity index.

From Croff's analyses (C4), it appears that the importance of ^{14}C waste management to the HTGR fuel cycle is already recognized. This problem is also being addressed in other countries. In Japan research is underway (M3) to incinerate the graphite fuel prisms with CO_2 rather than oxygen. The resulting CO is then to be catalytically decomposed into elemental carbon and CO_2, the CO_2 is then recycled to oxidize more graphite. In this way a pure carbon waste is produced, which is of smaller volume and lower solubility than $CaCO_3$. The method of disposing of this ^{14}C-contaminated waste has not been determined.

Definition of the requirements for ^{14}C waste disposal appears to be an important step which may significantly affect the choice of the HTGR fuel cycle.

9.2 Tritium (^3H)

Tritium formed in or released to the reactor coolant is a potential environmental contaminant of the reactor site, and tritium remaining with the discharge fuel is a potential contaminant of the reprocessing plant. In light water reactors the dominant source of tritium is from ternary fission. For a 1000 Mwe pressurized water reactor fission-product tritium is formed at the rate of 1.88×10^4 Ci/yr for uranium fueling and 2.47×10^4 Ci/yr with self-generated plutonium recycle. The estimated rate of formation of tritium in the reactor coolant is shown in Table 9.2. All of this coolant tritium is released to the environment at the reactor site, largely in the form of highly diluted HTO in liquid effluents.

In the HTGR the principal non-fission source of tritium is from $^6Li(n,\alpha)$ in lithium contaminants in the graphite and core matrix. Lithium concentrations of 0.01 to 1.2 ppm in HTGR graphite have been reported (G2, H4).

TABLE 9.2. Estimated Tritium Production in the Coolant of a 1000 Mwe Pressurized Water Reactor.

Source	Tritium production Ci/yr
$^{2}H(n,\gamma)$	2
$^{10}B(n,^{8}Be)T$	360
$^{6}Li(n,\alpha)$	34
$^{7}Li(n,\alpha)T$	4
Total from activation reactions	400
Fission-product tritium[a/]	188
TOTAL	588

[a/] Assumes fission-product tritium diffusing through fuel cladding or escaping through pin-hole cladding failures is equivalent to release of fission-product tritium from 1% of the fuel.

The tritium thus formed evidently diffuses to the coolant, so we shall estimate the average yearly production of ^3H in the coolant due to $^6Li(n,\alpha)$. At much lower concentrations the lithium is exposed homogeneously to the neutron flux. Because of its large thermal-neutron cross section, ^6Li is significantly depleted during the typical fuel irradiation time of 4 years. The average yearly production of ^3H from this reaction is then given by

$$<\dot{N}\lambda>_T = \frac{\lambda_T N_6^0 \phi\sigma_6}{T_R(\lambda_T - \phi\sigma_6)} \left[\frac{1-e^{-\phi\sigma_6 T_R}}{\phi\sigma_6 T_R} - \frac{1-e^{-\lambda_T T_R}}{\lambda_T T_R}\right] \qquad (9.7)$$

where λ_T = radioactive decay constant for ^3H

N_6^0 = initial number of atoms of ^6Li

σ_6 = (N,α) cross section of ^6Li

T_R = irradiation time of discharge fuel

For a core inventory of 362 Mg of C for a 1000 Mwe HTGR, and neglecting production of ^3H in the graphite reflector, we obtain

^3H from $^6L(n,\alpha)$ = 232 Ci/yr. for 0.01 ppm Li

= 2.79×10^4 Ci/yr. for 1.2 ppm Li

From calculations by Gainey (G2) of the $^{10}B(n,T)$ activation due to boron in HTGR control absorbers and burnable poisons, an additional 1250 Ci/yr of tritium is formed and diffuses to the coolant. Also reaching the HTGR coolant is about 0.5% of the fission-product tritium formed within the fuel particles, tritium formed in boron control absorber, and tritium formed by (n,p) reactions with ^3He impurities in the coolant. Because of the relatively large quantities of tritium thus formed, it is necessary to remove the tritium by reacting it with hot titanium in the continuous coolant clean-up system.

The small amounts (1.7×10^{-5}%) of ^3He present in underground sources of natural helium used for the HTGR coolant produces tritium by the reaction:

$$^3_2He + ^1_0n \longrightarrow ^1_1H + ^3_1H \qquad (9.8)$$

with a cross section of 5326 barns for 2200 m/sec neutrons and an effective cross section of 2800 barns at the HTGR operating temperature.

For an inventory of natural helium of 618 kg in the core of a 1000 Mwe HTGR (B12), ^3H is initially formed at the rate of about 8,020 Ci/yr. However, because of its large cross section, ^3H is rapidly depleted by neutron absorption. It is replaced by fresh helium introduced to make up for coolant leakage. If a fraction f_{He} of the coolant leaks from the coolant system per unit time, the steady state concentration X_{He-3} of ^3He within the reactor coolant can be calculated by

$$N^T_{He}\, f_{He}\, X^0_{He-3} = N^R_{He}\, X_{He-3}\, \phi\sigma_{He-3} + N^T_{He}\, X_{He-3}\, f_{He} \qquad (9.9)$$

where N^T_{He} = total inventory of helium in the coolant system

N^R_{He} = total inventory of helium within the reactor core

X^0_{He-3} = atom fraction of ^3He in natural helium $(1.7\text{x}10^{-7})$

Solving for X_{He-3}, we obtain

$$X_{He-3} = X^0_{He-3} \frac{1}{1 + \dfrac{N^R_{He}\, \phi\sigma_{He}}{N^T_{He}\, f_{He}}} \qquad (9.10)$$

From HTGR design data, it is estimated (B12) that

$$\frac{N^R_{He}}{N^T_{He}} = 0.09$$

$$f_{He} = 0.015/\text{yr}$$

For an effective $\bar{\sigma}_{He-3}$ = 2800 barns, and for $\phi = 1.2 \times 10^{14}/cm^2sec$, we obtain

$$X_{He-3} = 2.63 \times 10^{-9}$$

The resulting steady-state rate of production of tritium in the coolant from $^3He(n,p)$ is 124 Ci/yr.

In the CANDU heavy-water reactor the dominant source of tritium is the deuterium activation reaction. Data given for the Douglas Point Nuclear Power Station (C6) provide a basis for estimating the rate of production of tritium in the heavy water moderator and coolant:

electrical power = 203 Mwe
inventory of D_2O coolant in reactor core = 2.45×10^3g
average thermal neutron flux in coolant = $6.10 \times 10^{13}/cm^2sec$
inventory of D_2O moderator in reactor core = 7.18×10^7g
average thermal neutron flux in moderator = $1.01 \times 10^{14}/cm^2sec$
average $^2H(n,\gamma)$ cross section = 4.45×10^{-4} barns

The rate of production of ^3H in the D_2O is then:

$$(2.45 \times 10^3 \times 6.10 \times 10^{13} + 7.18 \times 10^7 \times 1.01 \times 10^{14}) \frac{gm}{cm^2sec} \times 4.45 \times 10^{-28}cm^2$$

$$\times \frac{6.02 \times 10^{23} \times 2 \text{ atoms}^2H}{20.02g\ D_2O} \times \frac{\ln 2}{12.3yr} \times \frac{Ci}{3.7 \times 10^{10}/sec} \times 0.8 = 2.42 \times 10^5 Ci/yr$$

For a 1000 Mwe CANDU power plant with the same reactor lattice and with the same ratio of D_2O in core inventory to uranium inventory as in the Douglas Point Reactor, the yearly production of tritium in the heavy water would be

$$\frac{1000}{203} \times 2.42 \times 10^5 = 1.19 \times 10^6 \text{ Ci/yr}$$

Because of this large rate of tritium generation it is necessary to operate a small isotope-separation unit to prevent the build-up of large concentrations of tritium in the heavy water. The losses of heavy water are kept small enough so that only a very small fraction of the tritium is released to the environment. The yearly release of tritium reported for the Douglas Point Station is typically about 4000 Ci/yr, which is about 0.2% of the allowable release (D4).

The amounts of tritium produced annually by these different reactors are summarized in Table 9.3.

TABLE 9.3 Summary of Tritium Production in Reactors

Reactor Type	PWR	PWR	CANDU HWR	HTGR
Fuel	U	U and self-generated Pu recycle	U	^{235}U, Th, and recycled U
Fission-product ^{3}H, Ci/yr	1.88x10^4	2.47x10^4	1.88x10^4	9.59x10^3
^{3}H in coolant, Ci/yr	5.88x10^2 [a]	6.47x10^2	1.19x10^6 [b]	1.65x10^3 [c] 2.93x10^4 [d]

[a] See Table 9.2

[b] D_2O coolant + D_2O moderator

[c] 0.01 ppm Li in C, 0.5% release of fission-product tritium

[d] 1.2 ppm Li in C, 0.5% release of fission-product tritium

9.3 Sulfur-35, Phosphorous-33, and Chlorine-36 in HTGR Fuel

The graphite fuel blocks of the HTGR reactor contain sulfur contaminant, which originates from the pitch used to form the fuel-rod matrix material. Neutron activation of the 4.22% ^{34}S in natural sulfur results in 88-day ^{35}S, according to the reaction:

$$^{34}_{16}S + ^{1}_{0}n \longrightarrow ^{35}_{16}S + ^{0}_{0}\gamma \qquad (9.11)$$

for which the 2200 m/sec cross section is 0.24 barns. Assuming that sulfur is present at 193 ppm in the HTGR fuel(H4), it is estimated that 215 Ci of ^{35}S are present in the fuel discharged yearly from a 1000 Mwe HTGR reactor, after 150 days of storage. In the HTGR reprocessing the stable and radioactive sulfur will volatilize to follow the carbon dioxide from graphite incineration. The radioactive sulfur is a potential environmental contaminant that must be recovered. The amount of ^{35}S activity is greater than that of ^{14}C, and the inhalation RCG is over an order of magnitude lower for ^{35}S. The stable sulfur may interfere chemically with some of the recovery processes in the off-gas system.

Natural sulfur also contains 0.76% ^{33}S, which undergoes (n,p) reactions to form 25-day ^{33}P, according to

$$^{33}_{16}S + ^{1}_{0}n \longrightarrow ^{33}_{15}P + ^{1}_{1}H \qquad (9.12)$$

with a 2200 m/sec cross section of 0.14 barns. The estimated activity of ^{33}P in the fuel discharged annually from a 1000 Mwe HTGR, after 150 days of storage, is 1.1 Ci.

Another volatile radionuclide formed in HTGR fuel is 3.1×10^{5}-yr ^{36}Cl, formed by neutron activation of chlorine contaminant in the fuel, according to the reaction:

$$^{35}_{17}Cl + ^{1}_{0}n \longrightarrow ^{36}_{17}Cl + ^{0}_{0}\gamma \qquad (9.13)$$

Natural chlorine contains 75.77% ^{35}Cl, for which the 2200 m/sec activation cross section is 43 barns. Assuming 3 ppm chlorine in the fabricated HTGR fuel(H4), the estimated yearly production of ^{36}Cl from a 1000 Mwe reactor is 1.02 Ci.

These additional radionuclides volatilized in HTGR fuel reprocessing are summarized in Table 9.4.

TABLE 9.4. Additional Volatile Radionuclides in HTGR Discharge Fuel (1000 Mwe ^{235}U-Th-fueled HTGR, 80% capacity factor, 150 days storage)

	Ci/yr
^{35}S	215
^{33}P	1.1
^{36}Cl	1.02

9.4 Non-Volatile Radionuclides Activated in Fuel Element Structure

Fuel elements discharged from pressurized water reactors also contain radionuclides formed by neutron activation in the Zircaloy cladding, stainless steel end fittings, and Inconel spacers. A typical three-year irradiation of the metallic structure produces the radionuclides listed in Table 9.5, calculated for fuel elements discharged from a light-water reactor and stored for 150 days (B10). Neutron capture in stable ^{94}Zr forms 65-day ^{95}Zr and its decay daughter, 35-day ^{95}Nb. The radioactivity produced is large, but it is still smaller than the radioactivity of these two nuclides formed as fission products. Other large contributors to the cladding radioactivity are ^{60}Co, resulting from neutron capture in stable ^{59}Co, and ^{51}Cr, ^{55}Fe, ^{58}Co, and ^{68}Ni.

After 10 years of decay there is still appreciable radioactivity remaining, so irradiated cladding must be treated as a long-lived radioactive waste. The only species which persist after about a thousand years of decay are 1.5x10^{6}-yr ^{93}Zr and 2.12x10^{5}-yr ^{99}Tc. The activity of ^{93}Zr in irradiated cladding is about the same as the activity of fission-product ^{93}Zr, but the activity of ^{99}Tc in cladding is about 1000 times less than the activity of fission-product ^{99}Tc.

The fast-breeder fuel cladding and structure, typically of 316 stainless steel, result in the radionuclides listed in Table 9.5 (B10). Since the structure is entirely an austenitic alloy, the most radioactive nuclides are ^{54}Mn, ^{55}Fe, and ^{60}Co.

The HTGR fuel contains no metallic structure, but impurities in the graphite fuel blocks result in the production of relatively small amounts of radioactive cobalt and nickel, as listed in Table 9.5 (H4). The total activity from metallic contaminants in HTGR fuel is considerably lower than that in the fuels from light-water and breeder reactors.

TABLE 9.5

Nonvolatile Radionuclides In Discharge Fuel From Neutron Activation

(1000 Mwe reactors, 80% capacity factor)

Reactor type[a] Fuel	Half Life	Activity in discharge fuel, Ci/yr PWR U_{235} (3.3% ^{235}U)	 HTGR ^{235}U, Th, and recycled U	 LMFBR U and recycled Pu
Beryllium 10	2.5×10^{6} yr		1.20×10^{-1}	
Sodium 22	2.60yr			5.16
Phosphorus 32	14.3day			23.7
33	25 day			3.16
Calcium 45	165 day	4.61×10^{-2}		
Scandium 46	83.9day	3.37×10^{1}		
Vanadium 49	330 day			7.04×10^{-1}
Chromium 51	27.8day	1.91×10^{4}		2.03×10^{4}
Manganese 54	303 day	4.79×10^{3}		1.74×10^{6}
Iron 55	2.6yr	4.89×10^{4}		1.30×10^{6}
59	45 day	6.17×10^{2}		1.47×10^{4}
Cobalt 58	71.3day	5.92×10^{4}	0.244	2.24×10^{6}
60	5.26yr	1.66×10^{5}	4.46×10^{-1}	3.22×10^{4}
Nickel 59	8×10^{4} yr	1.05×10^{2}	1.72	7.46×10^{1}
63	92 yr	1.56×10^{4}	2.28×10^{2}	2.37×10^{3}
Strontium 89	52 day	1.41×10^{2}		
Yttrium 91	58.8day	4.69×10^{2}		
Zirconium 93	1.5×10^{6} yr	2.81		
95	65 day	1.59×10^{5}		
Niobium 92m	10.16day			2.09×10^{-1}
93m	13.6yr	2.90×10^{-1}		4.86
95	35 day	2.96×10^{5}		4.88×10^{1}
Molybdenum 93	>100 yr	5.45×10^{-1}		7.46×10^{1}
Technetium 99	2.12×10^{5} yr	3.81×10^{-1}		7.25
Tin 117m	14.0day	1.96×10^{2}		
119m	250 day	4.31×10^{2}		
121m	76 yr	9.16		
123	125 day	5.30		
Antimony 124	60 day	2.28×10^{1}		
125	2.7yr	1.10×10^{3}		
Tellurium 125m	58 day	4.97×10^{2}		
TOTAL		7.72×10^{5}	2.61×10^{4}	5.33×10^{6}

[a] PWR = pressurized water reactor
HTGR - high-temperatue gas-cooled reactor
LMFBR = liquid-metal-cooled fast breeder reactor
Data are calculated for 150 days after discharge for PWR and HTGR, 60 days after discharge for LMFBR.

9.5 ^{232}U in Uranium Recovered From Irradiated Thorium

The results of several different calculations of the concentration of ^{232}U in uranium recovered from irradiated thorium and recycled uranium are summarized in Table 9.6. The data of Shapiro (S1) for a pressurized-water reactor are the same as the ^{232}U concentrations appearing in the recycled uranium of Table 3.5 and 3.7, wherein all uranium in the discharged fuel is assumed to be recycled, except for process losses. In the case of ^{235}U make-up the recycled uranium includes uranium bred from thorium as well as residual uranium from the ^{235}U make-up. These data are quoted for the fifth generation of irradiation, i.e. the build-up of ^{232}U has been followed through each generation consisting of a full irradiation exposure followed by reprocessing, uranium recovery, and fabrication of that recycled uranium with additional thorium for the next generation of irradiation.

^{232}U concentrations calculated by Arthur and quoted by Rainey (R1) for a PWR fueled with thorium and ^{233}U make-up are considerably higher than those calculated by Shapiro (S1); possibly because of the high initial ^{232}U concentration (1300 ppm) assumed by Arthur for the make-up ^{233}U. The Arthur-Rainey results indicate a 15% increase in the ^{232}U concentration if the initial thorium contains 100 ppm ^{230}Th.

Arthur's (A2) calculations for a PWR fueled with thorium and denatured uranium indicate far less isotopic concentration of ^{232}U in this fuel cycle, evidently because of the dilution by the denaturing ^{238}U (see Chapter 7).

Mann and Schenter's (M4) calculations for an oxide-fueled LMFBR with ^{233}U-^{232}Th core fuel indicate equilibrium ^{232}U concentrations near the ^{232}U concentrations predicted by Shapiro for the thorium-fueled PWR's.

TABLE 9.6. Summary of Calculations of ^{232}U in Recycled Uranium Recovered from Irradiated Thorium

Reference	Reactor	Fuel	assumed ^{230}Th in make-up Th, ppm	calculated ^{232}U in recycled U, ppm
Shapiro (S1)	PWR	thorium + 93% ^{235}U make-up, all U is recycled, near-equilibrium (5th generation)	0	2600
Shapiro (S1)	PWR	thorium + Pu, all U and Pu are recycled, near-equilibrium (5th generation)	0	2800
Rainey-Arthur (R1)	PWR	thorium + ^{233}U make-up, all U is recycled, near-equilibrium (5th generation)	0 100	4000 4600
Arthur (A2)	PWR	thorium + denatured U, ^{235}U make-up, near-equilibrium (5th generation)	0 85	260 316
		thorium + denatured U, ^{233}U make-up, near-equilibrium (5th generation)	0 85	512 563
General Atomic (H4)	HTGR	thorium + 93% ^{235}U make-up recycled bred uranium, near-equilibrium (2nd generation)	100	742
Mann and Schenter (M4)	LMFBR (oxide fuel)	10% ^{233}U + 90% ^{232}Th in core, 2-year irradiation, equilibrium	0	2760
		100% Th blanket, 3-year irradiation	0	86

10. Summary and Conclusions

The present commercial light-water reactors and the CANDU heavy-water reactors can be adapted to thorium fueling with very little modification in reactor design. For the thorium fueling in these reactors to be useful, a closed fuel cycle is required, involving the reprocessing of discharge fuel and recovery and recycle of fissile material. The reprocessing-refabrication technology for urania-thoria or urania-plutonia-thoria fuel is basically similar to that for urania-plutonia fuel, but more development and scale-up experience is required before the closed fuel cycle for thoria systems can be implemented.

The near-commerical HTGR is already designed for ^{235}U-Th fueling. However, reprocessing and refabrication operations for HTGR fuel differ in many respects from the present technological base established for urania-plutonia fuels. Considerably more development, beginning at the pilot-plant scale, is required.

Fueling LWR's and HTGR's with thorium and with plutonium recovered from uranium fuel discharged from LWR's is a logical way to introduce thorium fueling, but it achieves no better savings in uranium resources than recycling this plutonium as mixed-oxide urania-plutonia fuel in light-water-reactors. About 20% further savings in the reactor-lifetime uranium ore requirement is possible if the LWR or HTGR is fueled initially with thorium and 93% make-up ^{235}U, with recovered uranium to be recycled. This ore saving is calculated for first-generation reactors that must begin operating with no equilibrium fissile inventory in the reactor and fuel cycle. The time to reach equilibrium is relatively long in the uranium-thorium cycle.

The CANDU heavy-water reactor requires 40% less uranium ore when fueled with natural uranium and recycled self-generated plutonium than does the LWR with self-generated plutonium recycle. For a CANDU reactor started with natural uranium and converted to plutonium-thorium fueling as plutonium is recycled, the lifetime uranium ore requirement is less than half that of a uranium-fueled LWR with self-generated plutonium recycle. If started initially with 93% ^{235}U-Th, the CANDU lifetime uranium requirement is reduced to 39% of that of the uranium-fueled LWR with self-generated plutonium recycle. CANDU fuel elements must be modified to accomodate the higher burnups associated with these fissile-recycle fuel cycles. If started with natural uranium and converted to thorium fueling as self-generated plutonium is recycled, the CANDU reactor can reach self sustaining breeding, with a total uranium ore requirement within about 23 years, with a total uranium ore requirement 45% of the lifetime requirement of the uranium-fueled LWR with self-generated plutonium recycle. No additional uranium ore would be required for subsequent replacement thorium breeders.

The larger uranium ore savings possible with the CANDU reactor are a consequence of the relatively large conversion ratio of this reactor, which is 0.75 with uranium fueling, an average of 0.92 with the modes of high-burning thorium fueling analyzed in the present study, and 1.0 for low burnup thorium fueling with uranium recycle. Other studies indicate that with some lattice modification the CANDU conversion ratio can be further increased.

Improvements in conversion ratio of the PWR and HTGR, accompanied by further reduction in lifetime uranium ore requirements, are possible, but they appear to involve considerable design modification to achieve even the present conversion ratios of the CANDU reactor.

Given a reactor industry already based upon LWR's, the most direct and resource-effective approach to conserve uranium resources is to use plutonium from water reactors to start plutonium-uranium-fueled fast-breeders. Given a stockpile of depleted uranium as an already mined resource sufficient to fuel fast breeders for thousands of years at the end-of-century energy demand, the natural uranium resource required for the fast breeder is that attributable to supplying the start-up plutonium for the first-generation breeder. This ore for breeder startup represents a 32% increase in the lifetime uranium ore requirement for the light-water reactors producing the plutonium for the replacement breeders, assuming that these light-water reactors would otherwise operate with self-generated plutonium recycle. Plutonium start-up is the most resource-effective start-up approach for fast breeders. Thorium fueling with fast breeders offers no resource advantage, in the absence of special constraints that may be introduced by safeguards considerations.

Safeguards considerations have led to concerns about the recovery and utilization of plutonium in the power-reactor fuel cycle. The logic of the possible use of plutonium for nuclear explosives applies equally well to 93% ^{235}U. Therefore, until these concerns are resolved, cycles involving plutonium recycle or implementation of the Pu-Th or 93% ^{235}U-Th fuel-cycle alternatives might require that the reactors and reprocessing-refabrication operations involving these fuels be under special security control., such as location in specially controlled "international" centers.

These safeguards concerns have suggested the possibility that denatured uranium, i.e., uranium containing a low enough fissile concentration to be unsuitable for nuclear explosives, is sufficiently self-protected that reactors operating with such fuel can be safely dispersed as "national" reactors. The fuel discharged from these dispersed reactors will contain plutonium, but the plutonium in that form is assumed to be sufficiently self-protected by the intense radioactivity of the fission products. The discharge fuel would be shipped to the international center for reprocessing, and recovered plutonium would be consumed in plutonium-burning reactors colocated at the international center. From these analyses herein it is concluded that:

(a) Present 3% ^{235}U-^{238}U LWR fuels and natural uranium CANDU fuels are suitably denatured for such dispersed reactors. To obtain uranium-resource benefits from plutonium utilization, the power of plutonium-burner reactors at the international center must be an appreciable fraction of the total power of the nuclear power system. For LWR reactors, the ratio of international plutonium-burner power to the power of dispersed natural reactors is 0.4.

(b) Plutonium production can be suppressed, and the necessary relative power of the international plutonium-burning reactors reduced, by fueling the national reactors with thorium and denatured ^{233}U-^{235}U-^{238}U. Still using an international U-Pu-fueled PWR, fueling national reactors with thorium and denatured ^{233}U-^{235}U-^{238}U reduces the ratio of Pu-burning-reactor power to dispersed-reactor power to 0.14 for LWR dispersed reactors and to 0.07 for CANDU dispersed reactors. Further reduction is possible with Pu-Th-fueled plutonium burners.

(c) Similar combinations are possible with plutonium-thorium-fueled fast breeders, located at the international center, furnishing ^{233}U for the dispersed national reactors. However, this breeder power must be relatively large and the effective doubling time for breeder fissile-inventory is considerably lengthened, thereby decreasing the rate at which breeders can be introduced.

(d) Denatured ^{233}U-^{238}U, with a fissile concentration of about 12% ^{233}U, is relatively vulnerable to non-commercial isotopic enrichment to concentrations possible for explosives.

The total alpha activity of recycled plutonium in the uranium-plutonium fuel cycles is considerably greater than the alpha activity of recycled uranium in a uranium-thorium fuel cycle for the same reactor capacity. The higher-energy gammas from ^{232}U daughters accompanying recycled ^{233}U may require greater-shielding in fuel fabrication than in uranium-plutonium systems. The largest quantities of ^{232}U are calculated for uranium-thorium fuel in light-water lattices. Thorium recovered from irradiated fuel must be stored for many years for ^{228}Th decay before it can be recycled; 3 to 17 years are estimated for the reference HTGR U-Th fuel cycle.

The HTGR discharge fuel, whether from the U-Pu or U-Th fuel cycle, will contain relatively large quantities of ^{14}C diluted by a large amount of non-radioactive carbon from graphite incineration. The disposition of this long-lived solid waste is an environmental issue which warrants further study. The relatively large production of tritium in the CANDU heavy-water reactor is an environmental feature of this reactor, whether fueled with uranium or thorium.

It is the choice of fuel cycle, rather than the choice of the reactor, which has the greatest effect upon the long-term radioactivity and ingestion radio-toxicity of the high-level radioactive wastes. Differences in long-term radioactivity and toxicity are due more to differences in actinide composition and production, rather than to differences in the yields of fission products. In ^{235}U-Th fueling relatively little americium and curium are formed, and relatively little ^{239}Pu and ^{240}Pu appear later from the decay of americium and curium. Consequently, the ingestion toxicity of U-Th fuel-cycle wastes is relatively small during the period beginning at about 600 years after reprocessing, when ^{90}Sr and other fission products have decayed, until about 30,000 years. Relatively large quantities of ^{234}U and ^{238}Pu in the wastes from the U-Th fuel cycle result in a peak in ^{226}Ra radiotoxicity at 190,000 years of greater magnitude than in any of the other reprocessing fuel cycles and comparable to that in unreprocessed discharge uranium fuel.

With Pu-Th fueling relatively large quantities of Am, Cm, and Pu appear in the wastes. The long-term radiotoxicity due to these actinides in wastes is within an order of magnitude of the long-term radioactivity of the same radionuclides in unreprocessed discharged uranium fuel.

The long-term ingestion radiotoxicity of thorium mill tailings is less than that of uranium mill tailings in the ^{235}U-Th near-equilibrium fuel cycle, provided the natural thorium contains no contaminant ^{230}Th. If the natural thorium contains 100 ppm ^{230}Th, the ingestion toxicity of thorium mill tailings is increased to ten times that of the uranium mill tailings.

11. Acknowledgments

The authors express special appreciation for the guidance and assistance provided by Mr. Bruce Mann, Chief of the Evaluation Branch, Office of Radiation Programs, Las Vegas Facility, who was the EPA Project Officer for this study.

The study was supported in part by the Energy/Environment program of the EPA Office of Energy, Minerals, and Industry, Assistant Administrator for Research and Development, and in part by the University of California. Funding for computer time was provided by the EPA Office of Radiation Programs.

This final report reflects the many useful and constructive suggestions and comments resulting from reviews of an earlier draft by several agencies and individuals, including:

> DOE, Nuclear Power Development Division, Assistant Director for Fuel Cycle, Office of Waste Management
>
> EPA, Office of Radiation Programs, Technology Assessment, Division staff
>
> Dr. C.E. Till, Director Applied Physics Division, Argonne National Laboratory
>
> Oak Ridge National Laboratory Staff
>
> Dr. Bal Raj Sehgal, Program Manager Nuclear Safety and Analysis Department, Electric Power Research Institute
>
> Dr. Norton Shapiro, and Dr. R. A. Matzie, C-E Power Systems, Combustion Engineering, Inc.
>
> Professor S. Banerjee, Department of Engineering Physics, McMaster University, Ontario, Canada
>
> Mr. Mitsuru Maeda, Japan Atomic Energy Research Institute, Tokai, Japan. During his recent appointment as Research Associate at the University of California, Mr. Maeda reviewed many parts of the report and contributed directly to fuel-cycle calculations for Chapter VII.

The many sources of outside information, both through published work and by direct imput from individuals, are identified and credited in the list of references.

The authors are grateful for the help of Mrs. Sue Thur in preparing the manuscript and to Mrs. Edith Boyd, EPA, for proofreading and editorial assistance.

The contents of the final report, including interpretations and conclusions therein, remain the sole responsibility of the authors.

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13. Nomenclature

A	atomic weight of uranium isotope
E	fuel exposure
J	total interstage flow in ideal close-separation cascade
L	capacity factor
M	mass of isotope in fuel charged to reactor
N	total number of atoms of a radionuclide
Q	separative work
RCG	radioactivity concentration guide for ingestion, i.e., maximum permissible concentration in water
T_c	preprocessing cooling time
T_F	time elapsed between reprocessing and fabrication of recycled uranium-thorium fuel
T_R	fuel residence time in reactor
T_s	post-processing storage time for recovered thorium
x	atomic fraction of light isotope
α	ideal separation factor
β	fraction of recovered thorium to be recycled with bred uranium
η	overall thermal efficiency
λ	radioactive decay constant
ϕ	separation potential (Chapter 7), neutron flux (Chapter 9)
ψ	^{228}Th activity in irradiated thorium relative to ^{228}Th activity in natural thorium
σ	microscopic cross section
02	^{232}Th
08	^{228}Th
22	^{232}U

Appendix A: Storage Time for Thorium Recovered From HTGR Fuel Reprocessing

Thorium recovered from reprocessing irradiated thorium fuel may have to be stored prior to recycle to allow time for decay of 24.1-day ^{234}Th and 1.91-yr ^{228}Th, which are formed by the reactions discussed in Chapter 2. After the fuel is discharged, and prior to reprocessing, the ^{234}Th activity decreases with time. However, the activity of ^{228}Th may increase if ^{228}Th is not in secular equilibrium with ^{232}U at the time of fuel discharge. Although the total of the ^{228}Th and ^{234}Th activities decreases with time, the activity from ^{228}Th daughters is the most troublesome when chemically purified thorium is being refabricated. The highly energetic betas from both ^{228}Th and ^{234}Th chains give rise to large skin doses upon surface contact with separated thorium, but the highly energetic (2.6 MeV) gammas from the ^{228}Th decay chain can result in serious dose rates even with semi-remote fabrication techniques. Here we focus upon ^{228}Th, which controls the requirements for post-processing storage of recovered thorium.

When the separated thorium is to be eventually recycled and blended with low-activity uranium streams, such as make-up ^{235}U, the activity of ^{228}Th in recovered thorium after a preprocessing cooling time T_c and a post-processing storage time T_s is given by

$$(\lambda N)_{08} = \left[N_{22}(T_R)\lambda_{22}\left(1 - e^{-\lambda_{08}T_c}\right) + N_{08}(T_R)\lambda_{08}\, e^{-\lambda_{08}T_c}\right] e^{-\lambda_{08}T_s} \tag{A.1}$$

where

08 refers to properties of ^{228}Th
22 refers to properties of ^{232}U
$N(T_R)$ refers to the total quantity in the discharge fuel.

Thorium can be recycled for fabrication with low-activity uranium if the specific activity of ^{228}Th in the recycled thorium is no more than a factor Ψ greater than the specific activity of ^{228}Th in natural thorium,

$$(\lambda N)_{08} = \Psi(\lambda N)_{02} \quad \text{where 02 refers to properties of } {}^{232}\text{Th}. \tag{A.2}$$

Arnold$^{(A1)}$ suggests a value of $\psi = 5$ for thorium to avoid the requirement of semi-remote fabrication. Combining Eq. (A.1) and (A.2), we obtain

$$T_s = \frac{1}{\lambda_{08}} \ln \left\{ \frac{1}{\Psi} \left[\frac{N_{22}(T_R)\lambda_{22}}{N_{02}\lambda_{02}} \left(1 - e^{-\lambda_{08}T_c}\right) + \frac{N_{08}(T_R)\lambda_{08}}{N_{02}\lambda_{02}}\, e^{-\lambda_{08}T_c} \right] \right\} \tag{A.3}$$

For the reference HTGR reactor of Chapter 6 with discharge concentrations of $(\lambda N)_{22}/(\lambda N)_{02} = 4.05 \times 10^3$, $(\lambda N)_{08}/(\lambda N)_{02} = 1.70 \times 10^3$, $T_c = 150$ days, and $\tau = 5$, we obtain

$$T_s = 16.5 \text{ yr}$$

for thorium to be used when fabricating fuel with make-up ^{235}U. In the HTGR about two thirds of the thorium is used to fabricate fuel containing make-up or recycled uranium containing no ^{232}U, so about two thirds of the separated thorium would be subjected to the storage time estimated above.

For that portion of the separated thorium which is to be eventually recycled and blended with the recycled bred uranium, less time for thorium storage is necessary. A reasonable criterion is that the thorium be stored for a sufficient period such that its ^{228}Th activity is equal to the activity of ^{228}Th in the recycled uranium at the time of fabrication. Ignoring cross-over and process losses, the recycled bred uranium contains all of the ^{232}U which was present in the discharge thorium. If this recovered uranium has been stored for a time T_F prior to fuel fabrication, the activity of ^{228}Th in the uranium is

$$(\lambda_{08}N_{08})^U = N_{22}\lambda_{22}\left(1 - e^{-\lambda_{08}T_F}\right) \qquad \text{(A.4)}$$

Applying the above criterion, we equate the ^{228}Th activity in the bred uranium to the activity of ^{228}Th in the fraction β of the recovered thorium that is eventually to be recycled for fabrication with the bred uranium, i.e.,

$$(\lambda_{08}N_{08})^U = \beta(\lambda_{08}N_{08})^{Th} \qquad \text{(A.5)}$$

where $(\lambda_{08}N_{08})^{Th}$ is the activity of ^{228}Th in thorium after times T_c and T_s. Combining Eqs. (A.1), (A.4), and (A.5), we obtain

$$T_s = \frac{1}{\lambda_{08}} \ln\left\{\beta\,\frac{1 - 1 - \dfrac{N_{08}(T_R)\lambda_{08}}{N_{22}(T_R)\lambda_{22}}\; e^{-\lambda_{08}T_c}}{1 - e^{-\lambda_{08}T_F}}\right\} \qquad \text{(A.6)}$$

For the ^{235}U-Th-fueled reference HTGR reactor, $\beta = 0.36$. Assuming $T_C = 150$ days and $T_F = 60$ days, we obtain:

$$T_S = 3.1 \text{ yr}$$

for the recovered thorium to be used when fabricating fuel with bred ^{233}U. As the pre-fabrication time T_F of uranium storage increases, less time is required for thorium storage. For the parameters listed above if the recovered uranium is stored for 166 days before fabrication, the ^{228}Th activity in the uranium becomes equal to that of 36% of the separated thorium, so no additional time for thorium storage would then be required to meet the ^{228}Th criterion of Eq. (A.5).

Appendix B: Tables of Actinides in CANDU Fuel Cycles

NOTE: IN ALL APPENDIX B TABLES, α REFERS TO ALPHA-ACTIVITY, AND β REFERS TO BETA-ACTIVITY.

TABLE B.1 Actinides in the Fuel Charged To The Natural Uranium - Fueled CANDU Reactor (1000 Mwe, no Reprocessing[a])

		kg/yr	Ci/yr	weight %
Uranium[b]	235	9.075×10^{2}	1.946	0.715
	238	1.267×10^{5}	4.222×10^{1}	99.285
Total		1.276×10^{5}	$\alpha = 4.417\times10^{1}$	100.00

---

[a] 7.5 Mw-day/kg of U, 30.5% thermal efficiency, 80% capacity factor, equilibrium fuel cycle.

[b] ^{234}U is not included.

TABLE B.2 Actinides In The Fuel Discharged From The Natural Uranium-Fueled CANDU Reactor [a] (1000 Mwe, no Reprocessing [b])

		kg/yr	Ci/yr	weight%
Uranium[c]	235	2.233×10^{2}	4.788×10^{-1}	0.18
	236	1.003×10^{2}	6.361	0.08
	238	1.258×10^{5}	4.192×10^{1}	99.74
Total		1.261×10^{5}	$\alpha = 4.876\times10^{1}$	100.00
Plutonium[d]	238	5.312×10^{-1}	9.475×10^{3}	0.11
	239	3.201×10^{2}	1.963×10^{4}	66.28
	240	1.227×10^{2}	2.778×10^{4}	25.41
	241	3.086×10^{1}	3.470×10^{6}	6.39
	242	8.74	3.408	1.81
Total		4.829×10^{2}	$\alpha = 5.689\times10^{4}$ $\beta = 3.470\times10^{6}$	100.00

---

[a] immediately after discharge

[b] 7.5 Mw-day/kg U, 30.5% thermal efficiency, 80% capacity factor, equilibrium fuel cycle.

[c] ^{234}U, ^{237}U and ^{239}U are not included.

[d] ^{236}Pu and ^{243}Pu are not included.

TABLE B.3 Actinides In The Fuel Charged To The 1.2% ^{235}U-Fueled CANDU Reactor (1000 Mwe, no Reprocessing[a])

		kg/yr	Ci/yr	weight%
Uranium[b]	235	5.577×10^{2}	1.195	1.20
	238	4.592×10^{4}	1.530×10^{1}	98.80
Total		4.648×10^{4}	$\alpha = 1.650\times10^{1}$	100.00

---

[a] 21 Mw-day/kg U, 30.5% thermal efficiency, 80% capacity factor, near-equilibrium fuel cycle.

[b] ^{234}U is not included.

TABLE B.4 Actinides In The Fuel Discharged From The 1.2% ^{235}U-Fueled CANDU Reactor[a] (1000 Mwe, No Reprocessing[b])

		kg/yr	Ci/yr	weight%
Uranium[c]	235	3.496×10^{1}	7.474×10^{-2}	0.08
	236	7.563×10^{1}	4.796	0.17
	238	4.507×10^{4}	1.502×10^{1}	99.75
Total		4.518×10^{4}	$\alpha = 1.989\times10^{1}$	100.00
Plutonium[d]				
	238	1.748	3.080×10^{4}	0.70
	239	1.209×10^{2}	7.417×10^{3}	48.42
	240	8.604×10^{1}	1.949×10^{4}	34.45
	241	2.315×10^{1}	2.602×10^{6}	9.27
	242	1.788×10^{1}	6.978×10^{1}	7.16
Total		2.498×10^{2}	$\alpha = 5.778\times10^{4}$ $\beta = 2.602\times10^{6}$	100.00

---

[a] immediately after discharge

[b] 21 Mw-day/kg U. 30.5% thermal efficiency, 80% capacity factor, near-equilibrium fuel cycle.

[c] ^{234}U, ^{237}U and ^{239}U are not included.

[d] ^{236}Pu and ^{243}Pu are not included.

TABLE B.5 Actinides In the Fuel Charged To The U-Fueled CANDU with Self-Generated Pu Recycle (1000 Mwe, with Pu Recycle[a])

		kg/yr	Ci/yr	weight%
Uranium[b]	235	3.742×10^{2}	8.023×10^{-1}	0.715
	238	5.234×10^{4}	1.744×10^{1}	99.285
Total		5.271×10^{4}	$\alpha = 1.824\times10^{1}$	100.00
Plutonium[c]	239	1.706×10^{2}	1.046×10^{4}	35.68
	240	1.610×10^{2}	3.647×10^{4}	33.68
	241	4.109×10^{1}	4.620×10^{6}	8.59
	242	1.054×10^{2}	4.111×10^{2}	22.05
Total		4.781×10^{2}	$\alpha = 4.734\times10^{4}$ $\beta = 4.620\times10^{6}$	100.00

---

[a] 18 Mw-day/kg U + Pu, 30.5% thermal efficiency, 80% capacity factor, near-equilibrium fuel cycle.

[b] ^{234}U is not included.

[c] 150 days cooling of discharge fuel before reprocessing 0.5% loss in reprocessing, 0.5% loss in fabrication. ^{236}Pu and ^{238}Pu are not included.

TABLE B.6 Actinides in the Fuel Discharged From The U-Fueled CANDU with Self-Generated Pu Recycle [a] (1000 Mwe, with Pu Recycle[b])

		Kg/yr	Ci/yr	weight%
Uranium[c]	235	4.732x10^{1}	1.015x10^{-1}	0.09
	236	4.856x10^{1}	3.080	0.09
	238	5.159x10^{4}	1.719x10^{1}	99.82
Total		5.169x10^{4}	α =2.037x10^{1}	100.00
Plutonium[d]				
	238	1.974	3.450x10^{4}	0.40
	239	1.682x10^{2}	1.032x10^{4}	34.25
	240	1.625x10^{2}	3.681x10^{4}	33.10
	241	4.578x10^{1}	5.148x10^{6}	9.32
	242	1.126x10^{2}	4.392x10^{2}	22.93
Total		4.911x10^{2}	α =8.207x10^{4} β =5.148x10^{6}	100.00

---

a/ immediately after discharge

b/ 18 Mw-day/kg U+Pu, 30.5% thermal efficiency, 80% capacity factor, near-equilibrium fuel cycle.

c/ ^{234}U, ^{237}U and ^{239}U are not included.

d/ ^{236}Pu and ^{243}Pu are not included.

TABLE B.7 Actinides In The Fuel Charged To The ^{235}U-Th-Fueled CANDU Reactor (1000 Mwe, with U recycle [a])

	kg/yr	Ci/yr	weight%
Thorium 232	3.426×10^4	3.746	100.00
Total	3.426×10^4	$\alpha = 3.746$	100.00
Uranium[b]			
232	1.110×10^1	2.377×10^3	0.01
233	4.674×10^2	4.430×10^3	39.02
234	1.673×10^2	1.035×10^3	13.97
235	2.879×10^2	6.172×10^{-1}	24.03
236	1.923×10^2	1.220×10^1	16.05
238	8.286×10^1	2.761×10^{-2}	6.92
Total	1.198×10^3	$\alpha = 7.855 \times 10^3$	100.00

---

[a] 27 Mw-day/kg Th+U, 30.5% thermal efficiency, 80% capacity factor, near-equilibrium fuel cycle.

[b] 150 days cooling of discharged fuel before reprocessing. 0.5% loss in reprocessing, 0.5% loss in fabrication.

TABLE B.8 Actinides In The Fuel Discharged From the ^{235}U-Th-Fueled CANDU Reactor[a] (1000 Mwe, with U Recycle[b])

		kg/yr	Ci/yr	weight%
Thorium[c]	232	3.335×10^4	3.647	100.00
Total		3.335×10^4	$\alpha = 3.647$	100.00
Protactinium[d]	233	3.393×10^1	7.042×10^8	100.00
Total		3.393×10^1	$\beta = 7.042 \times 10^8$	100.00
Uranium[e]	232	1.41×10^{-1}	2.443×10^3	0.01
	233	4.359×10^2	4.131×10^3	44.88
	234	1.744×10^2	1.079×10^3	17.96
	235	7.842×10^1	1.681×10^{-1}	8.08
	236	2.102×10^2	1.333×10^1	21.64
	238	7.219×10^1	2.406×10^{-2}	7.43
Total		9.712×10^2	$\alpha = 7.667 \times 10^3$	100.00
Plutonium[f]	238	6.643	1.161×10^5	63.10
	239	2.176	1.335×10^2	20.67
	240	8.390×10^{-1}	1.900×10^2	7.97
	241	5.681×10^{-1}	6.388×10^4	5.39
	242	3.019×10^{-1}	1.177	2.87
Total		1.053×10^1	$\alpha = 1.164 \times 10^5$ $\beta = 6.388 \times 10^4$	100.00

---

[a] immediately after discharge

[b] 27 Mw-day/kg Th+U, 30.5% thermal efficiency, 80% capacity factor, near-equilibrium fuel cycle.

[c] trace quantities of other Th isotopes are not included.

[d] trace quantities of other Pa isotopes are not included.

[e] ^{237}U and ^{239}U are not included.

[f] ^{236}Pu and ^{243}Pu are not included.

TABLE B.9 Actinides In The Fuel Charged To The Pu-U-Th-Fueled CANDU Reactor (1000 Mwe, with U Recycle[a])

		kg/yr	Ci/yr	weight%
Thorium	232	3.445×10^{4}	3.767	100.00
Total		3.445×10^{4}	$\alpha = 3.767$	100.00
Uranium[b]	232	1.089×10^{-1}	2.332×10^{3}	0.02
	233	4.606×10^{2}	4.365×10^{3}	66.89
	234	1.577×10^{2}	9.760×10^{2}	22.90
	235	3.990×10^{1}	8.554×10^{-2}	5.79
	236	3.033×10^{1}	1.924	4.40
Total		6.886×10^{2}	$\alpha = 7.675\times10^{3}$	100.00
Plutonium[c]	239	2.092×10^{2}	1.283×10^{4}	66.36
	240	8.019×10^{1}	1.816×10^{4}	25.44
	241	2.014×10^{1}	2.265×10^{6}	6.39
	242	5.713	2.228×10^{1}	1.81
Total		3.152×10^{2}	$\alpha = 3.101\times10^{4}$ $\beta = 2.265\times10^{6}$	100.00

---

a/ 27 Mw-day/kg Th+U+Pu, 30.5% thermal efficiency, 80% capacity factor, near-equilibrium fuel cycle.

b/ 150 days cooling of discharge fuel before reprocessing.

c/ ^{236}Pu and ^{238}Pu are not included.

TABLE B.10 Actinides in the Fuel Discharged From The Pu-U-Th-Fueled CANDU Reactor[a] (1000 Mwe, with U Recycle[b])

		kg/yr	Ci/yr	weight%
Thorium[c]	232	3.356×10^{4}	3.670	100.00
Total		3.356×10^{4}	$\alpha=3.670$	100.00
Protactinium[d]	233	3.345×10^{1}	6.942×10^{8}	100.00
Total		3.345×10^{1}	$\beta=6.942\times10^{8}$	100.00
Uranium[e]	232	1.119×10^{-1}	2.396×10^{3}	0.02
	233	4.293×10^{2}	4.068×10^{3}	63.46
	234	1.651×10^{2}	1.022×10^{3}	24.41
	235	4.257×10^{1}	9.127×10^{-2}	6.29
	236	3.941×10^{1}	2.499	5.82
Total		6.765×10^{2}	$\alpha=7.489\times10^{3}$	100.00
Plutonium[f]	238	2.136	3.733×10^{4}	1.84
	239	1.637×10^{1}	1.004×10^{3}	14.08
	240	6.038×10^{1}	1.368×10^{4}	51.95
	241	1.919×10^{1}	2.158×10^{6}	16.51
	242	1.815×10^{1}	7.079×10^{1}	15.62
Total		1.162×10^{2}	$\alpha=5.208\times10^{4}$ $\beta=2.158\times10^{6}$	100.00

---

[a] immediately after discharge

[b] 27 Mw-day/kg Th+U+Pu, 30.5% thermal efficiency, 80% capacity factor, near-equilibrium fuel cycle.

[c] Trace quantities of other Th isotopes are not included.

[d] Trace quantities of other Pu isotopes are not included.

[e] ^{237}U is not included.

[f] ^{236}Pu and ^{243}Pu are not included.

Appendix C: Calculational Methods

1. Light-Water Reactors

Cycle-by-cycle burnup calculations by Shapiro, et al. (S1) for 1000 Mwe PWR power plant fuel cycles operating on both uranium and thorium fueling with segregated recycle were used to derive the material quantities for the fuel cycles. The lattice code "CEPAK" was used in doing the point (zero dimensional) reactor calculations. This computer code is a synthesis of a number of other codes: "FORM", "THERMOS", and "CINDER", where "FORM" is for the epithermal resonance and fast calculations on a homogenized cell, "THERMOS" calculates the thermal spectrum for a one-dimensional representation of the fuel cell, and "CINDER" does the fuel burnup calculations in a critical spectrum calculated by "THERMOS" and "FORM". The spectrum calculations were repeated prior to each burnup calculation to account for the spectrum effects of the depletion of the fuel isotopes and the build-up of fuel and fission product parasitic absorbers. The excess reactivity for leakage and control margin was assumed to be 4%.

The material quantities were scaled according to the power level and were corrected to a capacity factor of 0.8. The lifetime-average quantities shown in the mass flow sheets were calculated by accumulating the cycle-by-cycle quantities over the reactor lifetime. The equilibrium cycles were calculated from the data for the last reload designed for full burnup.

The computer code "ORIGEN" was used to calculate the radioactivity and toxicity of the high-level wastes. The initial actinide quantities in the high-level wastes were obtained from the discharge fuel concentrations from the "CEPAK" outputs.

Lifetime ore requirements were calculated by accumulating the ore requirements of each cycle over the reactor lifetime. For the first generation fuel cycles, the initial core inventory was also included. For those cases involving recycle of fissile material, and/or supply of fissile material recovered from fuel reprocessing, the reactor was assumed to be run on the slightly enriched (3%) uranium or on the ^{235}U-Th fuel cycle until sufficient fissile inventory was accumulated with the reactor and fuel cycle so that the reactor could then operate on the equilibrium fuel cycle.

2. High-Temperature Gas-Cooled Reactors

Data for the reference HTGR flow sheet were adapted from the detailed calculations of the concentrations of the nuclides in the various HTGR fuel streams published by General Atomic Co. (H4). In their calculations, the "GARGOYLE" code was used to calculate the flux spectrum and to perform the burnup and activation calculations in nine energy groups (five fast and four thermal). The core was represented as a point by using the core average nuclide concentration, and the "GARGOYLE" code was used to determine the core average neutron spectrum in each group with core leakage introduced as positive or negative contributions to the fission source in each group. The nine-group cross sections were collapsed from the ENDF/B-IV file by the "MICROX" code, which calculates the correct spectrum from the nuclide concentrations and lattice geometry.

All the numerical values were based upon the ninth fuel reload, loaded into the reactor at the beginning of the tenth year of operation, the last reload presented in the GA report. These GA data were calculated on the assumption of no cross-over between fissile and fertile streams in fuel reprocessing.

For the fuel compositions of the subsequent reloads, the effective one group cross sections deduced from the GA data were used. If we assume the effective one group cross sections are constant from reload to reload, then the discharge fuel concentrations of any nuclide can be expressed as linear combinations of the initial nuclides concentrations, and constants can be calculated from the ninth reload data and be used to calculate the discharge fuel concentrations for the later reloads.

It was found that the concentration of ^{236}U in the recycle bred uranium fuel does not reach equilibrium even during the lifetime of a second generation HTGR which has started up with the reactor and fuel cycle inventory of the first generation HTGR. Because uranium equilibrium occurs so late after the introduction of HTGR's, we chose to concentrate on the fuel cycle deduced from the General Atomic data from the ninth reload.

Because of a higher than normal amount of make-up ^{235}U charged into the reactor on the earlier (fourth) reload, which, after later discharge and reprocessing, is fabricated to form the first-recycle ^{235}U fuel of the ninth reload, so there is a considerable perturbation in the fuel charged to the ninth reload as compared with previous and subsequent reloads. Therefore, the data shown in the flow sheet were obtained by back extrapolation from the later reloads to make the discharge concentrations vary monotonically from one reload to another.

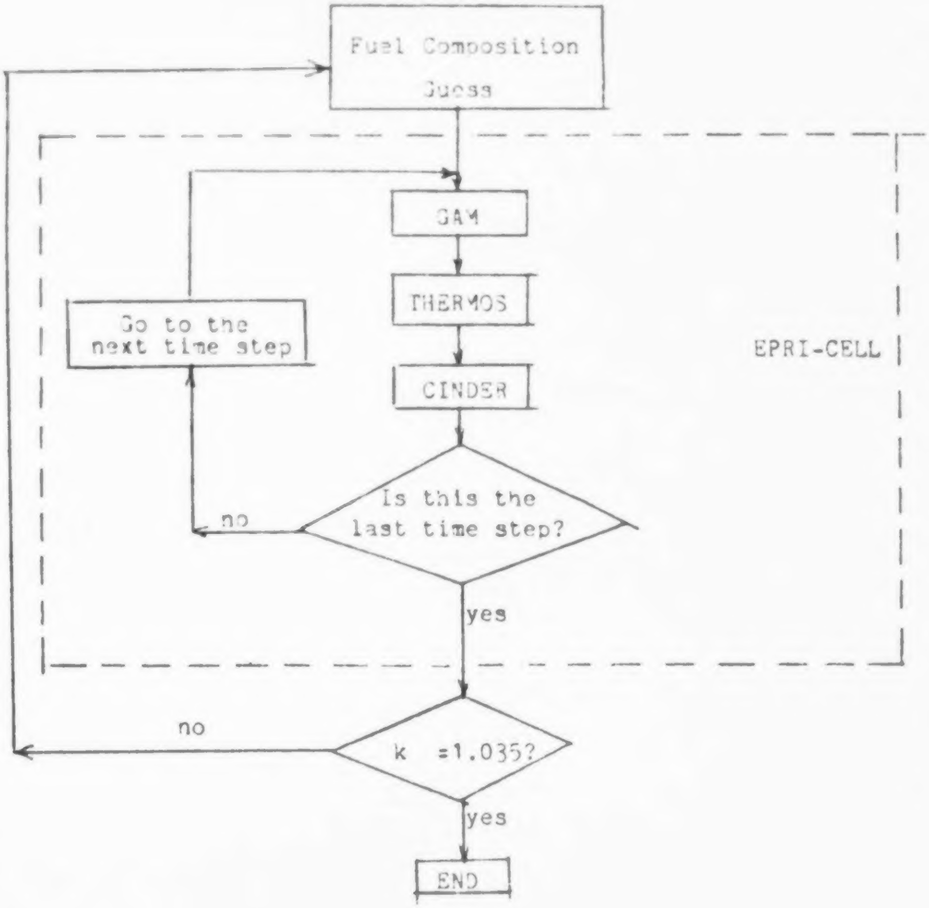
The effect of cross over on the fuel compositions was also calculated by using the constant effective one group cross sections method described above.

3. Heavy-Water Reactors

The goals for the calculation were to first determine the critical fuel composition at the beginning of cycle, and from this composition to determine the end-of-cycle discharge-fuel composition. The computer code "EPRI-CELL" (C7) was employed to do these calculations. "EPRI-CELL" is a computer code very similar to the "CEPAK" used in the PWR calculation by CE. It also has three built-in modules to calculate the space, energy and burnup dependent neutron spectrum within a cylindrical cell. "GAM" solves the Boltzmann equation to calculate the flux values for each of the 68 groups in the epithermal and the fast range. Nuclides can be specified in the input to receive heterogeneous resonance treatment; other nuclides in the cell will be treated homogeneously. "THERMOS" computes the thermal neutron spectrum (35 groups) as a function of position in a cell by solving numerically the integral transport equation with isotropic scattering. After the "GAM" and "THERMOS" calculations for one time step, the nuclide number densities, the cross sections of those nuclides included in the cell calculations, and the neutron spectrum are all passed on to "CINDER" to perform the depletion calculation. There are 20 depletion chains for 30 distinct heavy elements and 69 decay chains for 179 distinct fission products in the "CINDER" library. After each depletion calculation, the nuclide number densities are returned to "GAM" and "THERMOS" to perform the spectrum calculation for the next time step.

The heavy-water reactor cell used in the calculation is an equivalent cell to the actual 37-element CANDU fuel bundle, where there were 8 alternating fuel and coolant concentric rings followed by the coolant/calandria tubes and the moderator region.

The excess reactivity allowance for leakage, Xe override and control margin was assumed to be 3.5% (T1). Therefore, after each "EPRI-CELL" calculation, the infinite multiplication factor is calculated and tested. If it does not equal to 1.035, a new initial fuel composition is guessed and the whole calculation is repeated. The figure below shows the flow diagram of the calculation.


Fuel Composition Guess
GAM
THERMOS
CINDER
Go to the next time step
EPRI-CELL
Is this the last time step?
no
yes
no
k =1.035?
yes
END